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NBS SPECIAL PUBLICATION 331



# Radioactivity Calibration Standards

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## Radioactivity Calibration Standards

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International Conference of the American Nuclear Society  
Meeting on the Constructive Uses of Atomic Energy

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Edited by  
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## ABSTRACT

Experimental procedures and methods used in nine international radioactivity standardization laboratories are described by eight authors. The possibility of attaining accuracies of the order of 0.01% are discussed. The desirability of carrying out consistency checks of gamma-ray standards with time, using a  $4\pi\gamma$  ionization chamber is emphasized. The validity of radioactivity standards is examined. International comparisons organized by the Bureau International des Poids et Mesures are discussed. An analogue method of liquid-scintillation counting is described. Certain types of radioactivity standards are criticized. The fields of interest of the two Canadian laboratories are discussed. Details of the work in progress at the Boris Kidrič Institute and National Bureau of Standards are presented.

Key words: Alpha-particle standards; calibration consistency; coincidence counting; decay-scheme effects; efficiency tracing; gamma-ray standards; international comparisons; liquid-scintillation counting; radionuclide half-lives; radionuclide standards.

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## INTRODUCTION

The National Bureau of Standards was invited by the American Nuclear Society to organize a seminar on Radioactivity Calibration Standards at its November 1968 International Conference on the Constructive Uses of Atomic Energy in Washington, D.C.

These Proceedings contain the papers given at that seminar and the discussions that were generated by these papers. The discussions were recorded on tape and are presented verbatim, with a minimum of editing, in order to preserve their informality and vitality.

The delay in publication of just over a year, due to a variety of reasons ranging from initial production to final reproduction, is unfortunate. The importance and originality of this collection of papers on radioactivity standardization from many of the world's leading laboratories is such, however, as to mitigate against any harm due to delay, and most of the material and points of view presented herein are still unavailable elsewhere.

We would like to thank Mr. Oscar M. Bizzell and Dr. J. F. Black both for suggesting the seminar and for their support in organizing it.

W. B. Mann  
S. B. Garfinkel

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J. F. Black

I just wanted to take the opportunity to make a few remarks here this afternoon to introduce our program on the Radioactivity Calibration Standards to extend the appreciation of our division and society to the participants. This is one of the two special sessions which the Isotopes and Radiation Division is sponsoring here at the Washington International Meeting and is distinguished by the fact that first, it is one of the few sessions which consists entirely of invited papers and secondly, I think this session exemplifies the international air of the meeting in a better manner than most other sessions in view of the wide variety of the countries represented. Now I think we all realize that the organization of a meeting of this sort takes extensive time and effort from a man who has an international reputation which can draw the type of participation which has been achieved here and I think that we've been very fortunate in having such a man available as Chairman of the meeting in the person of Dr. Wilfrid Mann. I would like to extend the appreciation of not only the audience but the entire membership of the ANS Isotopes and Radiation Division to Dr. Mann for his efforts in organizing this fine program and I would also like to welcome the individual speakers whom he has invited and to extend our appreciation for the opportunity to hear about their work.

W. B. Mann

Thank you very much for your kind opening remarks and I'd just like to say very briefly how happy we are to welcome so many of our colleagues from abroad. The International Radioactive Metrology fraternity is a most pleasant one, and it has been one of my greatest joys to be able to work in this field.

I have been asked to read a paper for Dr. Hans Houtermans, but before I do that I'd like to say that we are going to try to publish the proceedings of this session and we do have a microphone and tape recorder going here, in order to get the discussion which can often be as interesting as the papers. The papers will be published and will be available I hope, and if anybody has a text and can give this to Mr. Garfinkel or to me I would deeply appreciate this, but the net result is that you will be able to skim over your papers and just touch on the high spots knowing that they will be published in their entirety, and if you exceed 20 minutes there will not be much time for discussion of your paper and as I say discussion can be very valuable.

I have glanced through Hans Houtermans' paper and I hope I can read it without detracting from it. In a covering letter, he said it did not have any figures or any jokes, and I am afraid I cannot supply any jokes. It's very short and I feel I have to read it. I cannot give you a brief resume. His paper is on "Aspects of Radionuclide Standardization in the I.A.E.A.:





# Aspects of Radionuclide Standardization in the IAEA

by

H. Houtermans

International Atomic Energy Agency, Laboratory

Vienna, Austria

One of the activities assigned to the laboratory of the IAEA in 1958 was the distribution of radionuclide standards. The responsible bodies of the Agency felt at that time that the supply of such standards was inadequate. Apparently, the fabrication of standard sources for all the numerous important radionuclides was neither scientifically very exciting nor interesting from a financial point of view. Therefore, this job was left to national standardizing institutes mainly, which were sometimes more inclined to improve calibration methods than to use their very limited staff for the distribution of many calibrated radionuclides. On the other hand, the performance of the necessary measurements just for a few samples of a not too frequently used radionuclide would, by any method of cost accounting, raise the price per sample to intolerable heights.

Since 1962, the IAEA laboratory has offered and distributed standards for a number of radionuclides. Because of its international clientele and by strict synchronization, the number of samples per distribution was relatively large, and thus the prices could be kept low.

Now it has been pointed out that during the last years the availability of radionuclide standards, perhaps due to a growing demand, has improved considerably, and standards of most radionuclides can be obtained from several different institutions. Therefore, it is appropriate to review the whole situation.

The intercomparisons between specialized laboratories, normally the "national standard" ones, organized by the BIPM, have shown agreement within 0.5% or better between the values of such institutions in some favorable cases. In principle, the activities of such standard sources from different laboratories may be expected to be consistent within these limits. But, for the overwhelming majority of radionuclides the situation is much less satisfying. Intercomparisons of standard sources from different suppliers have only been made in a few cases, probably, because even these relative measurements are by no means easy to perform to a precision definitively better than the overall uncertainty stated on the certificates.

Even in the most favorable case, gamma-ray emitters in solution form, possible errors due to weighing, not exactly reproducible conditions and non-linearity of the measuring device make it difficult for a non-specialized laboratory to decide that there is a discrepancy. Moreover, the samples are available nearly always at different times; uncertainties in half-lives, possible effects of small radionuclidic impurities and doubts about the long-term stability of the instrument response increase the possible errors.

It is probably for these reasons that the IAEA laboratory has, in all the past years, received practically no complaints about the activities given for the standardized samples. Because of the inherent sources of error,

selling radioactivity standards, especially short-lived ones, seems to be a fairly safe business.

It is now being considered that the IAEA laboratory may shift gradually its activity in the field of radionuclide standards from distribution to intercomparison by relative methods. There is no reason whatsoever that IAEA standards are "better" than those fabricated elsewhere, but IAEA as an international organization may be in a favorable position for registering and comparing calibration results obtained elsewhere.

At present, a scheme along the following lines, based on a proposal by Dr. W. B. Mann, is being discussed. It is assumed that the IAEA laboratory can preserve calibration results by means of some relative device such as a  $4\pi$  re-entrant ionization chamber for gamma-ray emitters. The quantity to be determined and preserved may be, in the case of a standardized solution of a special radionuclide, the quotient  $Q$  of the response of the instrument, when measuring a normalized sample of the solution, e.g. 2 grams, and the activity per gram according to the calibrating laboratory. For eliminating fluctuations of the instrument sensitivity, the sample can also be compared with a long-lived standard source. Then the essential quantity is  $A_e$ , i.e. the activity of the nuclide, which gives the same instrument response as the long-lived standard, when measured under standard conditions. Of course care must be taken to keep all conditions such as ampoule dimensions, absorption in the liquid etc. constant.

Institutions with practical experience in radionuclide standardization may now be invited to convey standardized samples in solution form of as many gamma-ray emitting nuclides as possible. On the basis of the reported value

for the activity per gram, IAEA will measure the equivalent activity  $A_e$ .

If two samples have been received of the same radionuclide from different laboratories, a list of the  $A_e$  values for this radionuclide will be started and be sent to those laboratories. They will now be asked to grant permission to include their result in further lists; otherwise their value will be kept confidential.

Laboratories sending samples later will receive their  $A_e$  value together with the list as it stands at this date; they will also be given the choice between inclusion in the list or withdrawing their result.

Copies of the list containing new values will be sent from time to time to all laboratories whose values are already included.

To increase the usefulness of the scheme, error estimation and other details to be included in the lists should be supplied by the calibrating institutions. A special form may be devised for obtaining uniformity.

One important point is the radionuclidic purity of the samples. At the beginning, IAEA will determine  $A_e$  under the assumption that impurities do not influence the results. At a later stage of the program, the radionuclidic purity of the samples may also be checked by IAEA in order to avoid inclusion of results for "impure" solutions in the list.

If this scheme is successful, each laboratory can compare its results with those of many other specialized institutions, irrespectively of the time the calibrations were performed. The IAEA laboratory will, of course, parti-

cipate itself, if possible even furnishing the first  $A_e$  values on the lists.

No interpretation of the results by IAEA is planned, but of course, each laboratory will draw its own conclusions. The immediate effect of the scheme will certainly be an increase in consistency of radionuclide standards, but an improvement of the accuracies can also be expected.

If the scheme shows its usefulness for gamma-ray emitters, it can be extended to other radionuclides also. For example, special proportional counters, windowless or with a thin window, can be used for solutions containing high- and medium-energy beta-ray emitters, liquid-scintillation counting for low-energy beta-ray emitters and electron-capture nuclides.

Many people think of a radioactive standard source, which has been calibrated by a relative method "only", automatically as one of a minor quality. This opinion is not justified and many unnecessary "absolute calibrations" are performed, not taking into account earlier results and those of the many other laboratories. If connected with radioactivity measurements, "absolute" just means "not depending on other standardizations", or more simply "starting from zero". If done properly such calibrations are clearly elaborate and time-consuming; they should only be performed occasionally for checking the relative method, or if one feels one can now do better than last time. For a standard, the only figure which matters is the overall uncertainty - "absolute" or "relative", indirect or more or less direct is of no importance to the user.

W. B. Mann

Is there any discussion? I don't propose to answer any questions, because I cannot, on behalf of Dr. Houtermans of the Agency. Maybe we should have some discussion of the subject. I think Dr. McNair was making rather the same point this morning about the preservation of measurements.

O. M. Bizzell

How does this compare with the early schemes the Bureau used? For example, I recall the matter of standardization of iodine-131 beginning about 1947. This seems to be somewhat similar to the approach that was taken in the standardization of that medically important radioisotope. Would you care to comment on that, Dr. Mann?

W. B. Mann

1947 was even before my time or that of Miss Cavallo who is here, but who might be able to comment. I suspect that much of the early work was done by  $4\pi\beta$  counting.

L. M. Cavallo

Yes, it was, but earlier measurements were made by determining the aluminum absorption curves for iodine-131 and for radium D + E and then comparing the responses at the extrapolated intercept for zero absorber.

O. M. Bizzell

I have reference, specifically, to the cross-comparisons of iodine-131 that George Manov did in late 1948 and in 1949. Dr. Manov worked in cooperation with many hospitals around

the country that were using iodine-131 for diagnosis and treatment. He found variations by factors of 3 to 4 in activity measurements. As a result of these intercomparisons, iodine-131 activities administered to patients were brought into much better agreement with NBS calibrations. The patient benefited tremendously and the medical effects of iodine-131 became much more predictable.

W. B. Mann

That may have been by the extrapolation method that Miss Cavallo has just mentioned. Actually I remember the intercomparison as I was at Chalk River at the time and we agreed also to participate. Only later on did we construct our " $4\pi\gamma$ " re-entrant ionization chamber at NBS and that is one of the most valuable devices that we ever had because you have a constant check on your previous measurements and all your past wealth of data is preserved.

A. McNair

I'd just like to comment that at the Radiochemical Centre we have been doing just this sort of thing for the past seven or eight years. All of our absolute measurements of gamma-emitting standards are checked against previous measurements using a re-entrant type of ionization chamber. We also do this for other people's standards, whenever these are available to us, particularly those obtained from the National Physical Laboratory in the United Kingdom. We have measurements on Agency standards as well. We have this wealth of preserved information going right back into the past, and as Dr. Houtermans' points out, you can reduce a number of errors in absolute standardization just by averaging over a long period of time. The sort of thing that I am thinking of are aliquotting and dilution errors in an abso-

lute standard, errors due to random background fluctuations, etc., which for any one absolute standard are systematic errors. In 20 or 30 absolute standards spread over a period of time these become random errors in the overall calibration of the ionization chamber. There are also other errors, like errors in assessing impurities in the material which may or may not become effective random errors over a long period of time. In principle I think it is quite feasible that one can make relative measurements with an accuracy approaching that of so-called absolute measurements in a number of cases.

W. B. Mann

Thank you Dr. McNair.

I remember too at the last meeting of the BIPM at Sévres that Dr. Campion had graphs from international comparisons showing the consistency of results from different laboratories over a period of years, 5 or 10 years. It's quite interesting. He was simply taking from reports of the BIPM and measurements the NPL has made on solutions that they had been distributing. Is there any more discussion on this paper? I think probably we had better go on because as I say I am in no position really to answer for Dr. Houtermans, and so I would like to call on M. Spagnol of Euratom to give his paper on "Some Aspects of Radioactivity Standardization at CBNM of Euratom."



# Some Aspects of Radioactivity Standardization

at the CBNM of Euratom

by

A. Spagnol

Central Bureau of Nuclear Measurements of Euratom

Geel, Belgium

Thank you, Professor Mann, for the kind invitation and the opportunity to talk here about the work of our Euratom laboratory at Geel in Belgium. The part of our work I would mainly like to speak about pertains to recent refined improvements in the accuracy of radioactivity determinations. This part is especially interesting, because, on the one hand, it is intimately connected to current problems in nuclear physics, and, on the other hand, throws some light at the present limits of accuracy in activity standardization. But first some words about our laboratory and its activities in general.

Our radionuclides laboratory is part of the CBNM, Central Bureau of Nuclear Measurements, which is responsible for standards and measurements in the nuclear energy field in the European Community. The main interest of CBNM (comprised of about 200 people) lies therefore with neutron standards and data and consequently an important part of the activities of its radionuclides laboratory is tied to such neutron objectives. The staff of our laboratory is kept

below the maximum number, which according to one of Parkinson's laws still allows efficient working and is high enough to allow extensive team work. Of the present 18 members, four are guests from different countries. The instrumental equipment of the group consists of more than 50, generally self-built, counting devices, about 20 of which are in current use. This includes two electromagnetic  $\beta$ -ray spectrometers, 2 Ge(Li)-detectors and 8 Si(Li)- and Si (Surface-barrier)- detectors. Our annual budget is about \$40,000, and the production rate of publications nearly 10 per year.

The work of our group can be subdivided into three parts, which absorb approximately equal amounts of our time: 1) direct neutron work, 2) standardization, 3) work for the improvement of the accuracy of radioactivity measurements. Typical examples of the first kind of work are an international intercomparison of threshold detectors irradiated in about 15 different reactors, which we are just finishing, or the accurate determination of the thermal-neutron activation cross section of Co-59. In the field of standardization we took part in all the useful BIPM intercomparisons, supply about 100 special standards per year to other laboratories, and give assistance to other groups in and outside the CBNM.

Our efforts towards the improvement of the accuracy of radioactivity measurements can again be subdivided into three parts: 1) development and improvement of methods, 2) accurate determination of nuclear and atomic constants involved in radioactive decay, mainly decay-scheme parameters, and 3) investigation of the physical effects, which limit the accuracy of radioactivity determinations, mainly effects of higher order. About 20 papers pertaining to the first point have been published in the last four years, ranging from  $\beta$ -counting using Cerenkov radiation to the application of the coincidence-counting method for the accurate determination of

internal conversion coefficients. Recent work concerns, among other minor investigations, the reduction of photon induced afterpulses in proportional counters and the investigation of the  $\gamma$ -sensitivity of  $\beta$ -counters in the  $4\pi\beta\text{-}\gamma$  coincidence technique. An interesting result is the confirmation of the very recently established dependence of the time interval between pulses and afterpulses in a proportional counter on the travel time of electrons between cathode and anode. Consequently we reduced the diameters of our cylindrical  $4\pi\beta$ -counters to about 2.5 cm and can now apply dead times of 0.4  $\mu\text{sec}$  with less than 0.05% afterpulse contribution to the counting rate. Another result of some interest is that the widely applied tungsten and molybdenum wires are less suitable anodes for proportional counters, because they rapidly deteriorate while stainless steel wires behave much more satisfactorily. With afterpulses eliminated, short dead times, non-deteriorating anodes, suitable choice of the cut-off energies and proper correction of the  $\gamma$ -sensitivity of the  $\beta$ -counter we hope that we shall soon be able to reach an accuracy near 0.01% in  $4\pi\beta\text{-}\gamma$  counting. Some effort has also recently been directed to the investigation of current-measuring methods of radioactivity determination, which might replace pulse-counting methods at the accuracy level of better than 0.01%.

Since the accuracy of radioactivity measurements is very often limited by the inaccuracy of several percent, with which most decay schemes are only known, much work has been devoted in the last years to the accurate determination of decay schemes and atomic constants. The schemes which we have re-investigated until now are shown in Figure 1. Only our results are indicated. Always as many as possible different methods have been used, for example, four for the determination of the  $\beta^+$ -branch in the Co-58 decay. The results on the Cs-137 decay and on the low intensity  $\beta$ -transi-

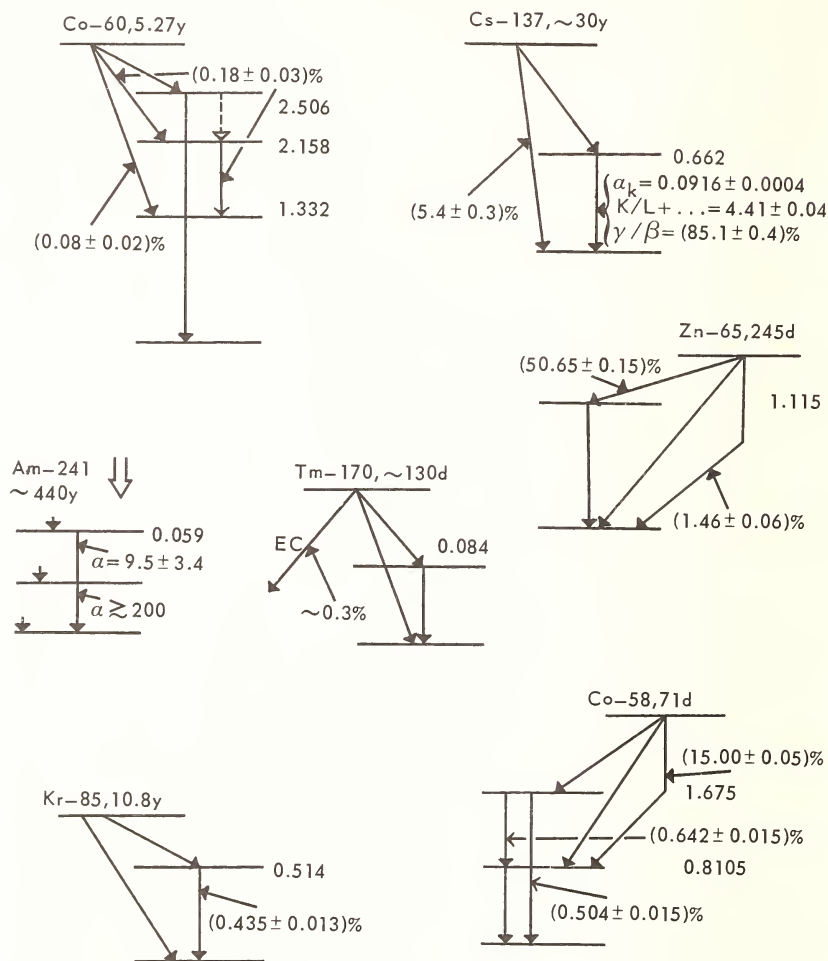


Figure 1.

tions in the Co-60 decay are perhaps interesting. The results on Zn-65 are not yet published, because we had a deviation of about 1% for the  $\gamma$ -branch intensity, when we used argon-methane instead of methane as counting gas in the  $4\pi\beta$ - $\gamma$  method. The argon-methane result is certainly wrong, because all other methods gave excellent agreement on the branching ratios. But we could not explain the difference until now and are still working on the problem. The results on Co-58 and Tm-170 are only preliminary, the final series of measurements are not yet finished. The measurements on the EC-branch in the Tm-170 decay are especially simple, since the branching is calculated only from spectra obtained with high resolution Si(Li)-detectors, which permit resolution of the K-X-rays of Er and Yb following electron capture or conversion after  $\beta$ -decay, respectively. Of the decay schemes which are at present under investigation that of Be-7 seems to be the most interesting one, also for neutron work. Atomic constants involved in radioactivity, which have been redetermined with an accuracy of better than 1%, are K-shell fluorescence yields of Cr after Mn-54 decay, Cu after Zn-65 decay and Cl after A-37 decay; those of several other nuclides are under investigation. Perhaps the most interesting of our results on nuclear constants are the conversion coefficients measured with our e-x-coincidence method to an accuracy of better than 0.5%. This accuracy could not be reached before and is therefore of some importance for the investigation of nuclear penetration effects in internal conversion. Finally it could be mentioned, that we redetermined about 20 half-lives with a rather high accuracy, including a microcalorimetric measurement on Tl-204, which was just finished. Many half-life measurements are still under way, the most important being those on the long living uranium isotopes.

The last point in our work for the improvement of accuracy in radioactivity determinations is the most interesting one. It is the investigation of effects of, in general, higher order intervening in radioactive decay. An uncomplete list of these effects, which often are subjects of recent studies in nuclear physics, is shown for a first orientation in Table 1, together with an indication of their order of magnitude. Some of them are under investigation in our laboratory. First results, for example, of an investigation of the ionization and simultaneous X-ray emission during  $\beta$ -decay of Pm-147 seem to confirm a recent theory, which predicts a rather great difference between total  $\beta$ -spectra and such taken in coincidence with X-rays. Also the theoretically interesting dependence of the ionization rate on the energy of the emitted  $\beta$ -particles is under investigation. With  $\beta^+$ -decays we have the complications connected with the annihilation. Problems are here the uncommon modes of annihilation, as one- or three-photon annihilation, directly or via positronium formation, and annihilation in flight. For the latter, figures can be found in the literature ranging from zero to several percent (relative to normal annihilation), a quite unsatisfactory situation. We estimated roughly the order of magnitude of this effect and try now to confirm our estimate by experiment. It is not impossible, that many  $EC/\beta^+$ -ratios measured so far have to be corrected for annihilation in flight. Another rather interesting problem is the intensity and energy distribution of high-energy electrons emitted during electron capture. This question is particularly important for Be-7, which as a "pure"  $\gamma$ -emitter is widely used for the determination of  $\gamma$ -efficiencies of  $\beta$ -counters. The total number of electrons with a continuous energy distribution accompanying electron capture amounts in this case probably to more than one percent, but the energy distribution of these electrons shows a rapid decrease with higher energy above about 50eV. Therefore it is not impossi-



the influence of the electron shell structure of the half-life, for example, of Be-7, which should be stable if it would be fourfold ionized. We are investigating the dependence of the half-life of Be-7 on temperature and chemical structure. First measurements at 300°C did not confirm an old theory that predicts a considerable dependence of the half-life on the temperature. Measurements at 2000°C as well as measurements with different Be-7 compounds are under way. These investigations on the half-life are closely connected to our L/K capture-ratio determinations on the same nuclide. Here, besides the investigation of environmental effects, the recently much discussed exchange and over-lap effects could be decisively checked. Our proportional-counter measurements, performed about three years ago using beryllium acetylacetonate at 300°C, failed, but new measurements using electrostatic analyzers are under way.

Finally it should be mentioned, that all our work of which I could only discuss examples, would not be possible without the fine spirit within the group, an extensive cooperation between its members and the excellent skill of our technicians.

Thank you!

W. B. Mann

Thank you, M. Spagnol. Would anybody like to ask M. Spagnol questions on his paper?

A. McNair

Two questions, M. Spagnol. When you speak of accuracies of the 0.01% in  $4\pi\beta\text{-}\gamma$  measurements, is this precision in measuring a particular source, or is this the overall accu-



racy of measuring the activity of a solution, including deposition on glassware of the radioactive material, the weighing uncertainties and all this sort of thing?

A. Spernol

I speak of the overall accuracy, including all effects, but I didn't mention in my talk all the other effects which should also be investigated such as impurity, and adsorption, and these effects include a lot of investigations that were not mentioned here.

A. McNair

So 0.01% is the accuracy of the final result for the solution?

A. Spernol

Yes, but one will not reach that in the first attempt; at the moment 0.01% is the order of magnitude which we hope to reach.

A. McNair

Do you take any special precautions about plating of radioactive materials on to vessels when dispensing or storing materials?

A. Spernol

We have alot of different precautions, for example, source preparation is made by an electro-balance which, as far as we know, has an accuracy better than one microgram but is difficult to check for droplets because there is no better

method. It was checked by standard weights and was better than one microgram. This is necessary for 0.01% overall accuracy.

A. McNair

Yes, well, I ask this question because we've noticed recently that for quite a large number of materials you can get absorption on vessel surfaces, ampoules and this sort of thing, of significant fractions of a microgram of material, so that if you're going to try to approach the 0.01% level of accuracy you're really going to have to have a lot of carrier or something else.

A. Spagnol

Yes, there is only a small range of activity and carrier where you can avoid this effect, but this must be checked in any case, this is quite clear.

A. McNair

That was one question. The second question: Have you investigated environmental effects on fluorescence yields?

A. Spagnol

Not sufficiently yet. What we did until now was to measure only K-shell fluorescence yields and environmental effects on K-shell fluorescence in  $^{54}\text{Mn}$  decay. The latter effect is certainly very low, because the nuclear charge is rather high. This environmental effect on fluorescence yields can only be expected to be considerable for the outer shells so we have had no results on it until now. But there are a lot of papers where the effect is described as x-ray-energy shifts.

A. McNair

Yes, I was thinking in particular of low-Z materials in the iron region, where if you look at the literature it seems that the published values of the fluorescence yield do depend on the state of the material when the measurement was made. Of course the accuracies of the measurements are often not very good, but it is very difficult to sort this sort of thing out.

A. Spernol

The effect of location on the fluorescence yields and their magnitudes is certainly below 0.1 or 0.2% and this cannot be measured at present.

W. B. Mann

I have asked this question before, when you reach 0.01% this seems to be our present limit of precision as you have to count, say, a microcurie, which is  $3.7 \times 10^4$  dps, for an hour, or  $3.6 \times 10^3$  seconds. That is a total of approximately  $10^8$  counts, giving a precision of  $10^4$ , but you are already counting for a long time and may run into problems of self absorption. You say you had already achieved this precision?

A. Spernol

Aiming for this. This is just one of the points which I mentioned. If you are to measure this number of counts you need low dead times. With normal proportional counters you cannot use dead times below, let us say, 5 or 10 microseconds due to the photon-induced after pulses but if you reduce this distance between the after pulses, by decreasing the diameter

of the counter, then you get this dead time. Our normal measurements last 12 to 24 hours and yield precisions much below 0.01%.

J. Silverman

Am I to understand that the state of chemical combination has no significant effect of fluorescence yields? There is, I believe, a known measurable effect on the half-life of beryllium-7 as a function of the state of chemical combination. I think Segré reported it some time ago.

A. Spagnol

Yes, this is a very unsatisfying situation. These theoretical investigations and measurements on beryllium-7 were done in the years 1948 to 1955, and the theory always gave influences on the chemical compound of the order of some percent. The first measurements showed this, but it was proved afterwards that this was wrong and the final measurements then showed that the effect is below 0.1%, two orders of magnitude below the theoretical prediction. But my belief is that not all of these experiments were very well done (especially the chemistry of the sources), so that the problem is not yet finally solved.

W. B. Mann

Thank you. The next paper is by Dr. Rytz of the International Bureau of Weights and Measures on "International Comparisons and Calibrations of Radionuclide Sources".

# International Comparisons of Calibrated Radionuclide

## Sources

by

A. Rytz

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Sèvres, France

Radioactivity designates both, the property of undergoing spontaneous nuclear transformation by emission of radiation (particles or photons) and more restrictedly, the rate of decay of these so-called radionuclides. Activity is measured in terms of disintegrations per unit time. The dimension being an inverse time, the same as that of a frequency, it is important to stress the basic difference between activity and frequency of recurrent events which stems from the fact that there is rigorously no connection between single decay events. This gives rise to a random distribution of rare events known under the name of the Poisson distribution. According to Poisson's law, the measurement of an activity  $A$  during seconds has a mean error of  $\sqrt{A/t}$ . Since this error is of the same nature as  $A$ , both magnitudes should have the dimension  $s^{-1}$ , rather than dps. Besides  $s^{-1}$ , the Ci is also in legitimate use ( $= 3.7 \times 10^{10} s^{-1}$ ) and is very nearly the activity of 1 g of  $^{226}\text{Ra}$ . Finally, activity may be expressed as the number of active atoms considered, multiplied by the decay constant  $\lambda$ , which is the decay probability per unit time. In all activity measurements we suppose that  $\lambda$  is a characteristic constant of the radionuclide. Since  $\lambda$  is a magnitude

which cannot be calculated exactly from other data, e.g. decay energy, it has to be determined experimentally with the aid of activity measurements. Thus there is an inevitable interdependence between half-life measurements and comparisons of activity measurements. In other terms: the radioactive impurities always set a limit of accuracy beyond which comparison of radioactivity is meaningless.

Activity measurements can directly be carried out in terms of  $s^{-1}$ . But in many cases, considerable self-absorption takes place and calls for correction terms which are so great that the measurement loses its absolute character. A super-purist might even argue that there is no really absolute activity measurement at all. I prefer a slightly less extreme attitude and would call absolute those measurements in which one counts single events, in an absolutely known solid angle, in terms of counts per second and where corrections are based on a sound theoretical basis. Uncertainties of corrections should not be much greater than the overall imprecision. Thus, ionization measurements should not be called absolute, but, under favorable conditions, proportional  $4\pi$  counting may give absolute results. The  $4\pi$  (PC)- $\gamma$  method, on the other hand, is considered to be the most reliable absolute method, provided it is applied to suitable radionuclides.

Scientific, medical, and industrial applications of radioactivity considerably increased the need of reliable absolute measurements of activity and availability of standard sources during the last decade. The smallness of the energy involved and the fact that part of it may be carried away by undetectable particles are the roots of the difficulties encountered in standardization of radionuclide sources.

Speaking about sources, we mostly think of solid sources, i.e. dry deposits on thin backing material. More generally, I should like to extend the notion of source to solutions of radionuclides from which solid sources may be prepared in a quantitative way. Liquid sources lend themselves better to the use of varying parameters and to shipping to distant laboratories. The quantity to be measured is, of course, activity per unit mass of solution.

If one carefully studies a certain method of standardization one can eliminate a great many systematic errors. If several laboratories compare their results obtained by measuring the same sources by the same method, some of the remaining systematic errors may further be discovered, i.e. eliminated. Yet there may still be systematic errors of some more hidden nature which escape detection unless different and independent methods of comparable precision are used. If this order of ideas and procedure is changed one takes the risk of increasing the effect of systematic errors.

The idea of international comparisons of sources and methods is not new. Discussion started in 1958 and three years later the decision was taken that the BIPM should extend its activity to the measurement of ionizing radiation and organize international comparisons of radionuclides. It was recognized that preparation and distribution was not the task of the BIPM, but that the aim was: getting coherent measurements between national standardizing laboratories. This obviously included the creation of a laboratory at the BIPM which can participate and take over the organization. A very ambitious program of comparisons was fixed and the aim was stated as follows: "An accuracy of  $\pm 1\%$  is desirable, going as far as  $0.1\%$  in special cases".

The practical work started in 1961 and during the first 18 months as many as six comparisons took place, including some rather difficult radionuclides. Table 1 gives a very rough account of the mutual agreement obtained. Further it illustrates what I just said regarding systematic errors. Nevertheless, I should like to mention that the last two comparisons enabled us to collect a great quantity of valuable information on many technical aspects of source preparation and counting.

TABLE I

Review of the first 7 international comparisons of radionuclides carried out under the auspices of the B.I.P.M.

Radio-nuclide	Reference date	Number of participants	Total spread (%)	Spread of selected labs.	Number of selected labs.
$^{32}\text{P}$	Jan. 61	16	6.0	2.0	13
$^{131}\text{I}$	Apr. 61	16	4.0	1.5	12
$^{198}\text{Au}$	Jan. 62	25	3.1	2.5 1.0	15 10
$^{60}\text{Co}$	Jan. 62	21	9.0	3.0	18
$^{204}\text{Tl}$	May 62	19	14.2	5.9	14
$^{35}\text{S}$	June 62	15	18.4	6.5	4

After a year's interruption a thoroughly prepared comparison of the  $4\pi\beta(\text{PC})-\gamma$  method, using  $^{60}\text{Co}$ , took place and turned out to be a real landmark in the history of international comparisons of radionuclides. Each of the 20 participating laboratories had to measure four solid sources of different activities which had already been measured by two experienced laboratories. In addition, a more usual comparison of a  $^{60}\text{Co}$  master solution was carried out.



The results can be summarized briefly as follows: For the first time, this comparison made it possible to discuss differences between results of the order of fractions of a percent instead of several percent. Between the results obtained by any two laboratories, the mean difference amounted to 0.6%, whereas the two pilot laboratories had a mean difference of 0.3% with a probability of 95%. Each participant had also to measure a thick source leading to a reduced  $\beta$ -efficiency and hence a considerable decay-scheme-dependent correction. It was found that this correction can be determined with sufficient accuracy. The results of the specific activity of the solution distributed gave a total spread of 1.4% which contrasts advantageously with that of preceding comparisons. Finally, there was some evidence of a systematic error due to the so-called extrapolation technique used in source weighing.

The solid sources had been most successfully prepared by electroplating gold-covered VYNS films by A. Spornol and his co-workers at Euratom (Belgium). These sources proved to be extremely stable and strong mechanically. At the BIPM we have sorted out four of them which we measure regularly by  $4\pi\beta$ - $\gamma$  coincidence counting. These repeated absolute measurements of the same sources may be considered as the creation of a "maintenance standard" of radioactivity. They raise a fundamental question: what should we trust more, the "conceptual standard", i.e. the  $4\pi\beta$ - $\gamma$  equipment or the "maintenance standard"? Before trying to answer, let us look at Figure 1. It represents a semi-logarithmic plot of 43 measurements of the sum of the 4 decay rates distributed over a little more than a half-life.

The first 17 results were obtained with the old equipment in the old building; most of the following results with the new equipment in the new laboratory. Different selections can

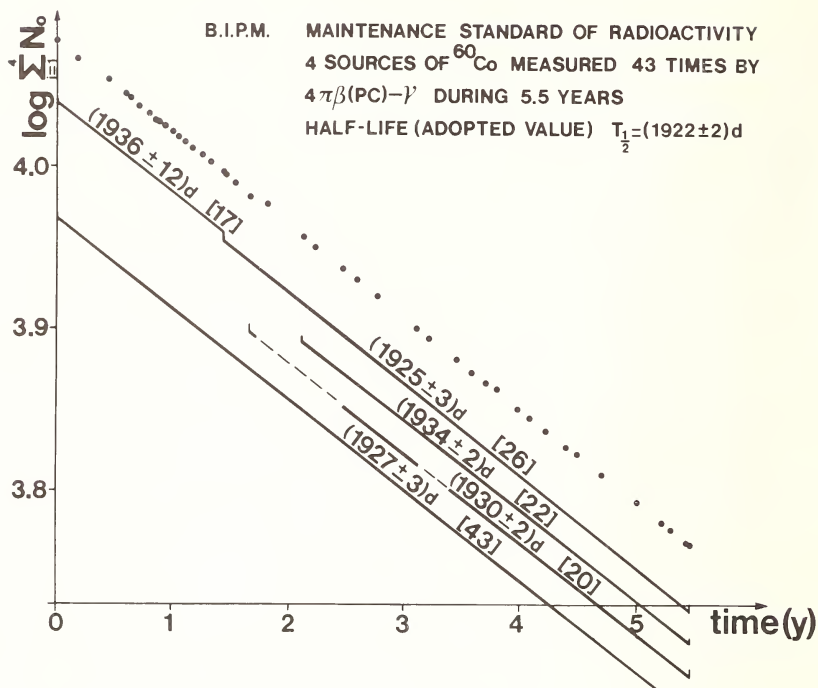


Figure 1.

therefore be made. I have calculated the half-life with four different sets of results, using a least squares fit to a straight line, assuming equal weights, which is not far from reality. All the half-lives thus obtained are slightly higher than the value found in the literature. Figure 2 shows the deviations from the best fit straight line. A  $\chi^2$  test confirms the visual impression that the 43 points do not seem to belong to a single Gaussian distribution. Even so, the mean deviation is only about 0.1%. We get even 0.05% with a more satisfactory  $\chi^2$  test if we take only the 20 last measurements obtained with the new equipment.

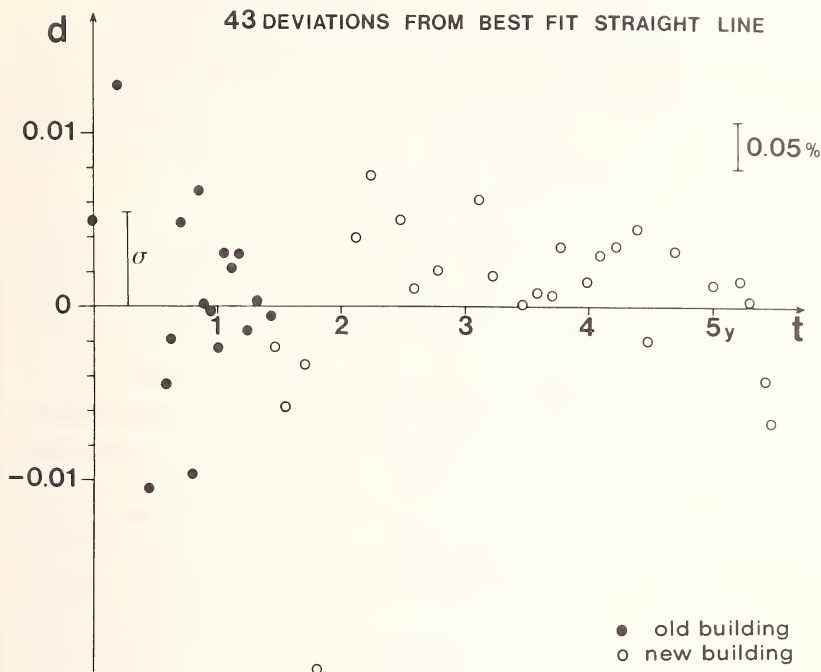


Figure 2. Deviations of 43 half-life measurements of  $^{60}\text{Co}$ .

It is not clear how we should interpret the deviations from the generally adopted half-life. Contamination can most certainly be ruled out. My personal feeling is that there is a very weak count-rate-dependent systematic error, e.g. in the dead-time correction. We are probably at the limit of the accuracy attainable with this method. The maintenance standard seems to be slightly more reliable than the conceptual one, but, of course, only during a limited time.

I shall not say much about the three subsequent international comparisons since they have also been reviewed at a panel meeting held in Vienna, two years ago. They concerned measurements of  $^{241}\text{Am}$ , an  $\alpha$ - $\gamma$  emitter,  $^{90}\text{Sr} + ^{90}\text{Y}$ , two pure  $\beta$ -emitters in equilibrium, and  $^{54}\text{Mn}$ , an electron capturer. The latter, unexpectedly, contained a large contamination of  $^{35}\text{S}$  which led to some difficulties.

In 1967,  $^{60}\text{Co}$  was again distributed in a large comparison which mainly aimed at the dilution and source-preparation techniques. Two solutions of different concentrations in an accurately known ratio were distributed, a concept which had already been used, though with less success, in the  $^{90}\text{Sr} + ^{90}\text{Y}$ , comparison. The participants were given very precise instructions as to how they should proceed in order to get comparable results. They had to prepare 28 sources from four dilutions of the strong solution and 14 sources of the undiluted weak one, and to measure them all by the  $4\pi\beta(\text{PC})-\gamma$  method following an exactly prescribed scheme.

About one half of the 25 participating laboratories have succeeded to determine the specific activity to within 0.1%, but there were also some results deviating by a percent or more. Dilution errors may have been the reason for this, in several cases.

The concentration ratio has been obtained correctly within 0.1% by one half of the participants. Four of them got it right within even 0.01%.

Systematic errors have often been greatly underestimated. In some cases the measurements can be shown to deviate significantly from a Poisson distribution or to be affected by a trend with the order of measurements or source preparation. The latter statistical analysis is rather complicated and will be explained in the report.

A preliminary comparison between 3 laboratories has been carried out before writing down the instructions and forms. This has been found to be extremely useful. Finally, the BIPM has worked out and distributed a detailed note on the frequently neglected buoyancy correction in weighing radioactive standard sources.

All the 10 preceding comparisons have been thoroughly discussed and described in great detail. The reports issued by BIPM may be obtained on request and will soon be completed by the report on the last  $^{60}\text{Co}$  comparison.

Summarizing, we see that a great amount of work has been invested in 11 very different comparisons. Many illusions have been destroyed, for hundreds of sources have been prepared and measured without any positive result. On the other hand, a wealth of useful information has been collected. A definite improvement of standardizing techniques has been reached. Many laboratories, especially the less advanced ones, have come to know their capabilities and their position with respect to others. Source preparation, dilution, and  $4\pi\beta\text{-}\gamma$  coincidence counting have been unified considerably. On the whole the work has been quite successful and worth the big effort made.

The success of comparisons of the kind described will always depend on the interest with which they are undertaken by the participants who may belong to one of two different groups. The first group includes laboratories with great experience in this field and for whom comparisons represent only a small part of their activity. They must squeeze them into an already heavily loaded standardization program. The second group is formed by laboratories with medium or little experience, wishing to improve their technique and test their equipment. For these, comparisons are welcome opportunities

to estimate the value of their work. Laboratories of the first type consider comparisons more or less as a sacrifice which they take upon themselves for the community's sake. For the second group comparisons may be of great help provided advanced laboratories take part as well. However, comparisons are neither the only nor even the best way of helping less advanced laboratories to improve their techniques. In many cases, a few calibrated sources or a standardized solution would be more efficient, more rapid and much simpler.

The investment of time and money in a comparison is always considerable on the organizer's side as well as on that of the participants.

Carrying out a comparison without a well defined aim or without sufficient interest on the part of the participants would, therefore, be a waste of man-hours and money. We know from experience that it is often very difficult, if not impossible, to compare results obtained by different methods, especially when they are of different levels of accuracy. On the other hand, if we confine ourselves to a single method, systematic errors may be carried along, without the possibility of detecting them.

A compromise between both concepts is necessary to get an undistorted view with regard to systematic errors. At this moment, the BIPM does not have definite plans for future comparisons. Better knowledge of special needs and interests of the advanced standardization laboratories will have to be considered and more discussion is needed before a decision can be taken. I very much hope that this discussion will start during this meeting.

Note added in proof

Figure 1 contains results which might be misleading as far as the half-life of  $^{60}\text{Co}$  is concerned. After a careful review of all the data and addition of five more recent measurements, we preferred to reject the 17 early measurements and some of the following ones because of lacking coherence in the measuring conditions. Thus only 21 measurements distributed over 3.3 years are retained. A least-squares fit using weights inversely proportional to the square of the experimental standard errors gives a value of  $(1927 \pm 2)$  days or  $(5.276 \pm 0.006)$  years, where the error stated is purely random (one standard deviation). We estimate that the systematic error should not exceed twice this amount.

W. B. Mann

Thank you, Dr. Rytz. Are there any questions?

Perhaps I might just make a comment rather than ask a question. I'm not quite sure whether our laboratory is among the advanced laboratories or not so advanced laboratories, but Miss Cavallo recently said to me that there has not been a single international intercomparison in which we have not learned something, so we are very grateful for the stimulus of these intercomparisons. Are there any questions?

R. S. Caswell

I'm a little concerned by this statement that there are no plans for the future for intercomparisons. Is there a mechanism that will review this? Is there some point in time when a decision will be made concerning possible future intercomparisons?

A. Rytz

I think we will decide this very soon. Even before the end of the year.

W. B. Mann

Are there any other questions for Dr. Rytz? If not, thank you very much, Dr. Rytz. It was a very interesting talk.

The next paper is to be given by Dr. McNair of the Radiochemical Centre in England. It is entitled "Standardization of Radioactive Solutions by Efficiency-Tracing Methods Using a Liquid-Scintillation Counter".



# Standardization of Radionuclides by Efficiency-Tracing

## Methods using a Liquid-Scintillation Counter

by

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### 1. Introduction

Radionuclides which emit only low-energy  $\beta$  radiation are notoriously difficult to standardize because coincidence methods cannot be applied directly to such nuclides. Gas-counting methods can be used for certain members of the group such as tritium, carbon-14 and sulphur-35, but these methods, though accurate, are rather difficult and tedious for routine standardization work. The simple  $4\pi\beta$  method of counting solid sources prepared quantitatively from a solution is too inaccurate to be of real value, because of wide variations in the self-absorption of the sources and the impossibility of making a direct experimental determination of the efficiency with which electrons from the sources are counted.

Some of the advantages of coincidence methods can be extended to the standardization of low-energy  $\beta$  emitters by a technique first proposed by Merritt et al. in 1960 (1,2). In this "efficiency-tracing" technique a suitable  $\beta$ - $\gamma$  emitting radionuclide is added to the "pure"  $\beta$  emitter and the relationship between the number of  $\beta$  particles of the pure  $\beta$  emit-

ter detected in the counter and the efficiency with which the  $\beta$  particles of the "tracer" radionuclide are detected (measured by coincidence methods) is determined experimentally. Rather sophisticated experimental work by Williams et al (3) has shown that this relationship is not, in general, linear but by working to high precision a meaningful curve can be fitted in the observable region and extrapolated to 100% tracer efficiency to give a figure which is presumed to be the true disintegration rate of the pure  $\beta$  emitter. Work, using different tracers and the same pure  $\beta$  emitter, has shown small, but apparently reproducible, systematic differences between results obtained with different tracers (4). Baerg (5) has considered the efficiency tracing method theoretically and has shown that the validity of the extrapolation depends on an implicit assumption that the counting efficiency depends only on the parameter being altered to change the efficiency. To get a closer approximation to a one-parameter system he prefers to change the efficiency of his sources by adding external absorbers instead of employing the more usual procedure of measuring a large number of sources containing different amounts of inactive carrier to give a range of counting efficiencies. Many of the difficulties that arise with efficiency-tracing measurements using solid sources are probably caused by the measurement techniques not satisfying Baerg's "one variable parameter" criterion. The most usual cause of the difficulties is generally ascribed to non-uniform and variable distribution of the "traced" and "tracer" radionuclides in the sources causing a variation in the relative efficiencies with which the  $\beta$  particles from the two radionuclides are detected. For this reason at the Radiochemical Centre we decided to look at the possibility of using a liquid scintillation counter for absolute standardization work.

In principle there are a number of attractive features in using a liquid-scintillation counter with the radionuclide

dissolved in the scintillant as the electron detector in standardization work. The source preparation should be simple and reproducible. There is no solid source to absorb the radiations emitted from the decaying nuclei, and a uniform mixture of radionuclides should be obtainable for efficiency-tracing measurements. A liquid-scintillation counter has the added advantage that it is a reasonable spectrometer for electrons of moderate energies and the counting efficiency can be varied in an uncomplicated manner merely by altering the amplification or external discriminator levels, to give low-energy discrimination.

The main disadvantage of a scintillation counter is the relatively low efficiency with which the energy of the primary ionizing particle is converted into a useful electrical signal. As a rough rule of thumb, counting efficiencies obtained for  $\beta$  emitters in a liquid-scintillation counter are not greatly different from those obtained in counting solid sources of the same radionuclides in a proportional counter. The difference is that the main loss of efficiency in a proportional counter is the result of electron absorption in the source, whereas in a liquid-scintillation counter the loss of signals occurs in the statistical variations in the processes of photon and, particularly, photoelectron production. These processes may be treated mathematically or, as we have done at the Radiochemical Centre, by designing an analogue of the liquid-scintillation counter that is close enough to reality to permit us to make a measurement of the efficiency of the counter which is accurate enough for standardization work. The principles and design of the system were described in 1966, in a paper presented at the IAEA Symposium on Standardization of Radionuclides (6). What follows is a brief description of the system before going on to discuss the results.

## 2. The Analogue System

The simplest and most accurate analogue system would use as much of the electronic equipment of the real scintillation counter as possible, and all that was done was to replace the bottle of scintillant by a pulsed light source which simulated in intensity and intensity distribution the scintillations produced by the interactions of  $\beta$  particles in the scintillant.

A cathode-ray tube with an aluminized "killed-phosphor" screen with a decay time of less than 0.1  $\mu$ sec. proved to be a satisfactory light source. Flashes of constant intensity are produced by pulsing the tube with 200 nanosecond pulses at the grid and these flashes illuminate the phototube cathode after passing through an aperture, a series of diffusers and filters and, finally, reflecting from a diffuse 45° reflector replacing the sample bottle. The intensity of the flashes is modulated by moving a shutter across the aperture. The position of the shutter is controlled by a cam and lever arrangement (Figure 1). Any desired modulation of the intensity of the flashes can be achieved by using a suitably shaped rotating cam to move the shutter across the aperture.

The instant at which each flash is produced is known so that the output pulse, if any, can be identified and the efficiency of the system measured under any desired conditions.

In order to simulate the scintillations produced in a bottle of scintillant by the  $\beta$  particles from a dissolved radionuclide, the shape of the cam must incorporate:

- A. The shape of the energy spectrum of the  $\beta$  particles. This is derived from Fermi theory.

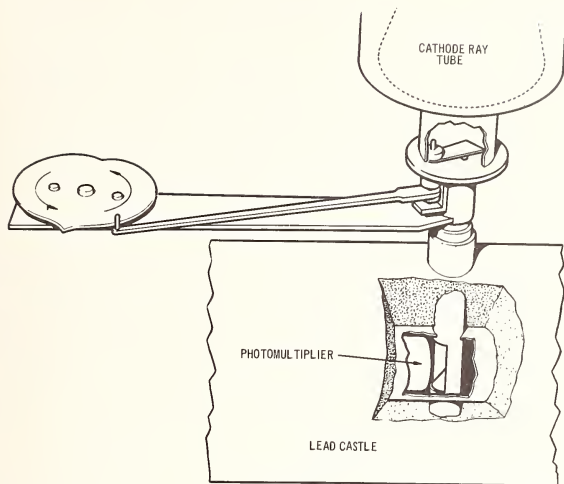


Figure 1.

B. The relationship between the efficiency of the scintillant and the energy of the exciting electron. The relationship published by Horrocks is used (7,8).

C. The relationship between light transmission and radius of the cam. This was obtained by using the cathode-ray tube as a continuous light source and measuring the current from the photo-tube as the lever was moved.

D. The energy range of the system. This is the energy of a particle which will produce a scintillation of the same mean intensity (at the photocathode) as the flashes reaching the photocathode when the shutter is fully open.

### 3. Preliminary Tests of the Analogue System

The first tests of the system were made to see how closely the pulse-height spectrum from simulated niobium-95 compared with the pulse-height spectrum obtained from a bottle of scintillant containing the radionuclide in solution. The intensity of the light flashes was adjusted so that the efficiency measured using the analogue system was the same as the real efficiency (measured by  $\beta$ - $\gamma$  coincidence methods) under particular conditions of gain and discrimination. Then the conditions were altered in the same way for both real and simulated spectra. Small systematic divergences between the two spectra were observed and similar results were obtained with real and simulated cobalt-60. However, there are a number of factors which, either singly or in combination, can be expected to create differences between the pulse-height spectra produced by the scintillation counter and its analogue. Most of these factors are likely to distort the real and simulated spectra of different radionuclides in the same way. The use of an efficiency-tracing type of measurement is therefore likely to minimize the distorting effects, because in this technique the spectrum of one  $\beta$  emitter is, in effect, compared with the spectrum from another  $\beta$  emitter, and the simulated spectra are also compared.

The most direct way of testing this is to measure the relationship between the efficiencies with which the  $\beta$  particles from a mixture of two radionuclides in the scintillant are detected, and compare this relationship with that measured using the simulated spectra in the analogue system. In order to make an unequivocal determination of the efficiency of detecting real  $\beta$  particles, suitable  $\beta$ - $\gamma$  emitting radionuclides were used, and the efficiencies were measured by coincidence techniques. Unfortunately, there are relatively few  $\beta$ - $\gamma$  emitters with decay schemes which permit the selection of

a  $\beta$  spectrum with the correct shape and in the desired energy range. The most suitable from the physical point of view are niobium-95, mercury-203 and cobalt-60. Measurements have been made on mixtures of niobium-95 and cobalt-60 in scintillant, and the results compared with the predictions of the analogue system are shown in Figure 2. The predictions of the analogue system are seen to be in good agreement with the measurements on the real radionuclides. Small divergencies between the real and simulated spectra observed at very low counting efficiencies may be caused by spectrum distortion due to wall effects in the bottle of scintillant.

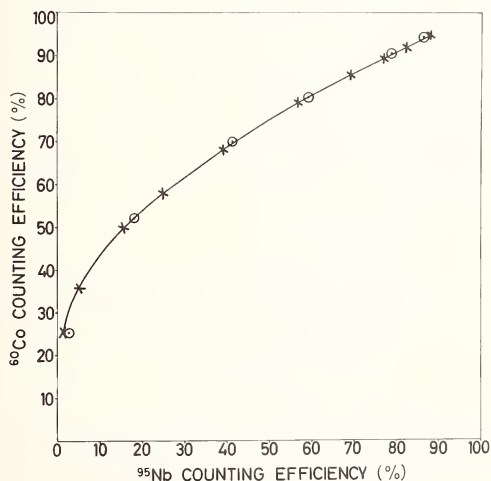


Figure 2.

#### 4. Measurements on Pure $\beta$ Emitters

Measurements of the activity of low-energy pure  $\beta$  emitters cannot of themselves provide unambiguous proof that the system gives correct results. This can only be implied, by comparing the results with measurements of the activity of the same materials by independent methods.

For the purpose of this illustration I shall concentrate on measurements that have been made on materials that have been independently standardized, usually at other laboratories. In particular, I shall discuss measurements on two solutions of sulphur-35 (end-point energy 167 keV) standardized at the National Physical Laboratory, a sample of n-hexadecane labeled with carbon-14 (end-point energy 159 keV) standardized by the National Bureau of Standards, Washington, and a sample of carbon-14 n-hexadecane measured at the Radiochemical Centre but by a direct comparison against NBS-standardized sodium carbonate-Cl4.

The procedure adopted was to obtain the relationship between  $\epsilon_\beta$ , the efficiency of detecting the  $\beta$  emitter, and  $\epsilon_{\beta_Y}$ , the efficiency of detecting the  $\beta$  particles from the tracer nuclide, using the analogue system. Then, using a mixture of the radionuclides in scintillant and varying the conditions of measurement, values of  $N_\beta$ , the number of  $\beta$  particles detected from the pure  $\beta$  emitter, and the corresponding efficiency with which the  $\beta$  particles from the tracer were detected (measured by coincidence methods) were obtained experimentally. The values of  $N_\beta$  divided by the corresponding values of  $\epsilon_\beta$  give what should be the disintegration rate of the pure  $\beta$  emitter in the scintillant. Clearly  $\frac{N_\beta}{\epsilon_\beta}$  must be constant irrespective of the value of  $\epsilon_{\beta_Y}$  at which the measurements are



made. This is a necessary though not a sufficient condition for the system to be valid for measuring a particular pair of radionuclides. Some examples of the results that have been obtained are shown in Figures 3-5 and a summary of the results of the measurements on the sulphur-35 and carbon-14 materials is given in Table 1.

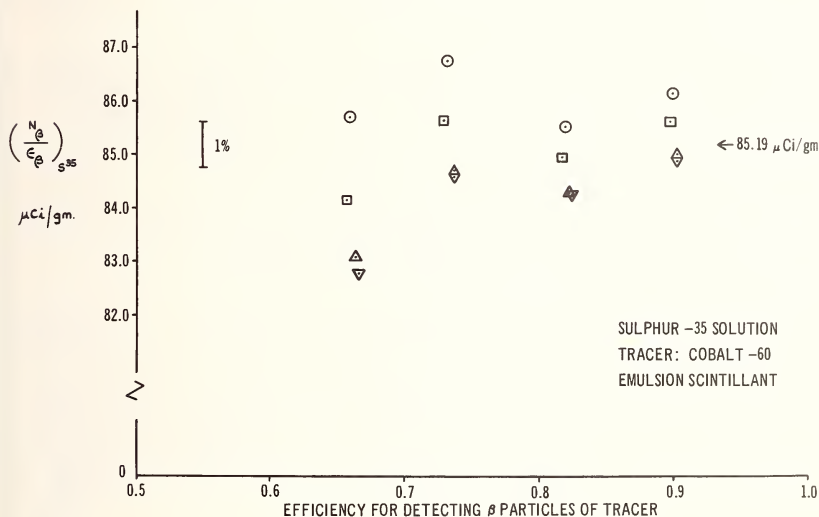


Figure 3.

It is clear from the table that very good agreement is obtained between measurements made using the analogue system and the results obtained by other laboratories. (It is worth noting that different methods of measurement were used at the other standardizing laboratories. For the sulphur-35 measurements, NPL used solid source-efficiency tracing methods, fitting a second-order polynomial to the results and extrapolating on the fitted curve. For the first of the two results quoted, three different tracers were used but the third result was quoted to lower statistical accuracy than the first two. The

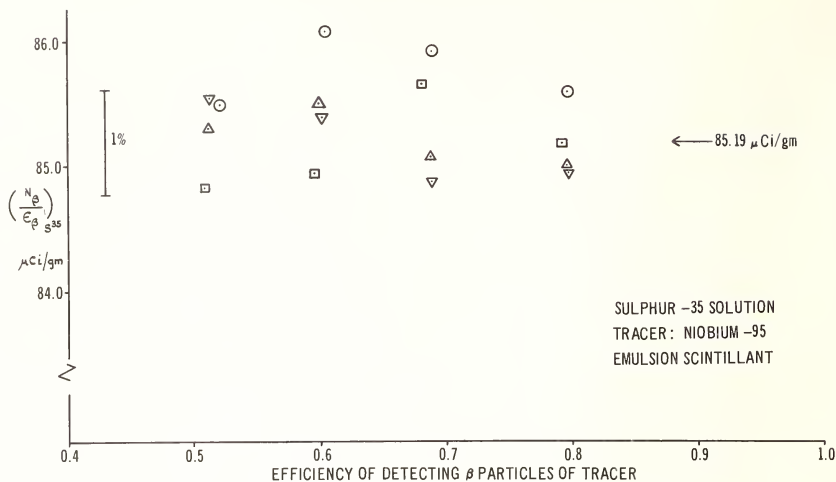


Figure 4.

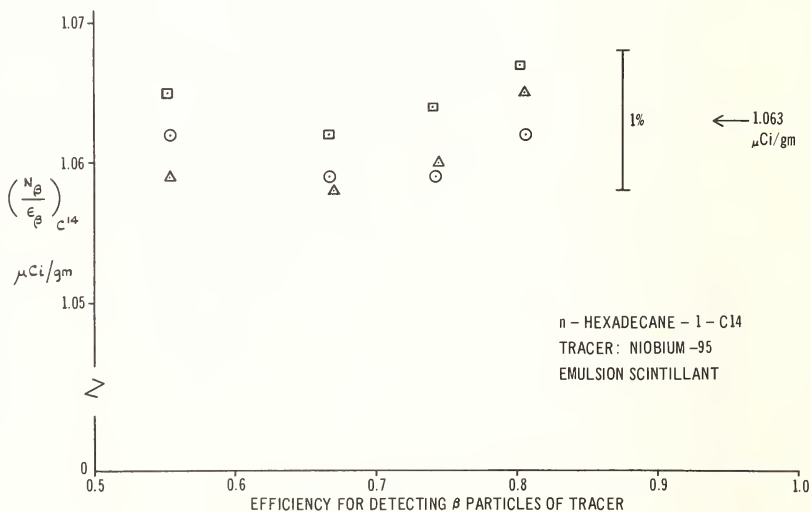


Figure 5.

Table 1 Results of Comparison Measurements

Radionuclide Measured	Tracer Used	Result from analogue system* ( $\mu\text{Ci/gm}$ )	Comparison Result ( $\mu\text{Ci/gm}$ )	Comparison Laboratory
Sulphur-35	Cobalt-60	404.5 $\pm$ 0.4	406.91 ) 401.98 ) 394.46 )	NPL
Sulphur-35	Cobalt-60	85.22 $\pm$ 0.21 )	85.19 $\pm$ 3.1	NPL
	Niobium-95	85.35 $\pm$ 0.12 )		
Carbon-14	Niobium-95	0.783 $\pm$ 0.002	0.781 $\pm$ 0.016	RCC, by direct comparison with NBS sodium carbonate C-1
Carbon-14	Niobium	1.062 $\pm$ 0.001	1.063 $\pm$ 0.010	NBS

\*The errors quoted for measurements on the analogue system are the 1 $\sigma$  statistical counting errors only. The errors on the comparison results are the overall errors quoted by the standardizing laboratory.

NBS measurement of hexadecane was achieved by oxidizing the hexadecane to carbon dioxide, preparing sodium carbonate from the carbon dioxide and comparing the activity of the sodium carbonate directly with sodium carbonate standards.) This good agreement, together with the results already discussed for  $\beta$ - $\gamma$  emitters goes a long way to convincing us that the system provides an analogue of the real scintillations which is probably quite as accurate as is required for efficiency-tracing measurements when the  $\beta$  spectra of the tracer and traced radionuclides are not too dissimilar in end-point energy. Calcium-45 (end-point energy 254 keV) has also been measured by the analogue technique with apparently satisfactory results.

## 5. Estimation of Residual Systematic Errors in the System

Having designed a system to measure the activity of low-energy pure  $\beta$  emitters it is important to be able to make a realistic assessment of the accuracy of the results obtained using the system. Estimating the random errors in the analogue-system measurements is fairly straight-forward, but as usual, the main problem is to estimate the effects on the results of systematic imperfections. So far, the only parameters that have been investigated have been the effects of inaccuracies in setting up the intensity of the light flasher for the analogue system, errors in measuring the light transmission law of the shutter, and the effects of uncertainties in the assumed end-point energies of a few of the  $\beta$  spectra.

The correct flasher intensity is set up by measuring the efficiency for detecting the  $\beta$  particles from the tracer under specific counting conditions and then varying the intensity of the flasher until the analogue system reproduces this efficiency for the simulated radionuclide. Then the relationship between the efficiencies of detecting tracer and traced  $\beta$  particles is measured using the analogue system. This was then repeated using a flash intensity only 50% of the "correct" value and the change in the analogue-measured efficiency for detecting sulphur-35  $\beta$  particles (when traced with cobalt-60) was 0.3% of  $\epsilon_\beta$  at  $\epsilon_\beta = 0.81$ , 0.4% of  $\epsilon_\beta$  at  $\epsilon_\beta = 0.64$  and 1.3% of  $\epsilon_\beta$  at  $\epsilon_\beta = 0.53$ . Tests show that the flasher intensity can certainly be set to within 10% of the correct value.

The effects of inaccuracies in the end-point energies of the  $\beta$  spectra act on the system in a uni-directional sense and are not compensated by the use of an efficiency-tracing technique. In order to investigate this (and other) parameters of the system a stepped cam was designed. With this, the probability of detecting a light flash as a function of

its intensity can be measured. The results of a measurement of this kind can then be combined with the other relevant data by computer to give the appropriate relative counting efficiency curves. Measurements have shown that a 1% change in the assumed end-point energy of calcium-45 causes about 1% change in  $\frac{N_{\beta}}{\epsilon_{\beta}}$  when  $\epsilon_{\beta}$  is 0.54 but only 0.2% change in  $\frac{N_{\beta}}{\epsilon_{\beta}}$  when  $\epsilon_{\beta} = 0.87$ .

For sulphur-35 the corresponding figures are a 1.5% change in  $\frac{N_{\beta}}{\epsilon_{\beta}}$  when  $\epsilon_{\beta}$  is 0.34 and 0.3% change when  $\epsilon_{\beta}$  is 0.80.

The stepped cam has also been used to investigate the effect of changes in the law of transmission of light through the shutter. Two versions of the relationship which differed by amounts comparable with the measurement errors were investigated but the effect on  $\frac{N_{\beta}}{\epsilon_{\beta}}$  for sulphur-35 was very small at  $\epsilon_{\beta} = 0.34$ . A more extreme variation in which the mid-point transmission of light was assumed to be only 18.6% compared with the measured 30.2% was also tried. This altered  $\frac{N_{\beta}}{\epsilon_{\beta}}$  for calcium-45 by about 0.2% at  $\epsilon_{\beta} = 0.54$ .

These results again show the value of using an efficiency-tracing measurement to reduce the significance of variations in many parameters of the system. Clearly, though, the investigations of systematic errors point out the value of making measurements in the analogue scintillation system at counting efficiencies that are as high as possible, consistent with not counting spurious events, in order to reduce the effects of systematic imperfections.

## 6. Conclusions

Investigations of the behaviour of the liquid-scintillation-counter system have shown that it is feasible to design an analogue of the system which can provide accurate enough results for standardizing a number of low-energy pure  $\beta$  emitters by efficiency-tracing measurements.

It is particularly noteworthy that, unlike other forms of the efficiency-tracing technique, the results are obtained without having to make an unknown extrapolation outside the observable region of efficiencies.

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K. D. George

In some of your previous slides, I recall one on hexadecane, you had some points that were triangles and some that were squares and consistent differences between these families. What were the differences between these triangles versus the squares?

A. McNair

These were just different samples of tracer plus hexadecane dissolved up in liquid scintillant.

K. D. George

Different Samples?

A. McNair

Different samples, yes. All the same material but different aliquots.

W. B. Mann

Any more questions? If not, thank you very much Dr. McNair, for your very interesting paper.

The next paper we had a little difficulty in translating the title. It seems to have come out as a misprint in this program too. I think it's "Validité des étalons radioactifs", but when we looked up "validité" in the dictionary we found it meant both "validity" and "availability" and either word would have done all right! So we await Mr. Le Gallic to tell us which it was.



# Validity of Radioactive Standards

by

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Standard radioactive products are essential to users to enable them to quantify their research work on the various applications of radionuclides. Further this is the logical conclusion to the radioactive metrological studies carried out by the national laboratories specializing in this particular field. Therefore, not only must radioactivity standards be developed and delivered but the measure of their validity must also be determined.

The standards may be divided into two classes: primary and secondary.

The primary standard is determined by a direct method of activity measurements, without any intermediate handling, e.g., a solid source measured by the  $4\pi\beta$  coincidence method, or a  $\gamma$  solution determined by calorimetry.

The secondary standard which results either from a direct measurement of activity on an aliquot, or from an indirect activity measurement, e.g., standard solutions calibrated, as from sources measured in  $4\pi\beta$ , or  $\gamma$  sources measured in a previously calibrated ionization chamber, gas samples calibrated by the compensated-counter method.

The secondary standards are the most used by far and this is why we shall study their validity first, but not without first exploding a myth.

### 1. The False Standards

I shall deal very quickly with this aspect of the question which, from the scientific angle, is of no great interest but which nevertheless is worth mentioning because this practice is growing.

The manufacturers of radiological protection measuring instruments have to ascertain their reliability and to do so they use reference sources. This is quite in order of course, but to keep abreast with rival firms it is essential for them to have competitive efficiency, and hence they blithely transform their reference source into an activity standard. Here are two examples of this:

#### A. Standard source for radiological protection used to determine surface contamination

These sources are prepared by depositing a known amount of a standard solution on to a solid surface, stainless steel for instance. The efficiency of the detector is then determined, and owing to backscatter of the  $\beta$  rays this is obviously increased from 10% to 30%.

#### B. Standard source for a liquid-scintillation counter

The industry making such equipment is a most flourishing one with a very promising future owing to the importance of these instruments

in medical and biological research. But efficiency is a fundamental criterion of choice for the user, so this is how high efficiency is obtained:

1. By taking a solution that has really been calibrated by a national laboratory, for instance some tritiated toluene,
2. By placing the optimum quantity of the tritiated-toluene-scintillator mixture in the customary counting vial, taking care to use only toluene triple-distilled over sodium,
3. By pumping out, then removing all traces of oxygen in the scintillating solution by purging with an inert gas, and then
4. By sealing in vacuo, taking care also to cover the stopper of the flask with, say a  $\text{TiO}_2$ -base reflecting paint.

Providing all this is done for  $^3\text{H}$ , an efficiency of 30% to 40% greater will be obtained than that of the user who is content to utilize classical scintillators, in air and without a reflector.

Such processes involve the legal liability of the manufacturers but they also involve the moral liability of the national laboratories who unwittingly serve as cover. Thus the radiological protection instruments which I mentioned a few moments ago are delivered with a secondary standard from the Saclay laboratory of radioactivity metrology and some liquid-scintillation counters with a secondary standard of the National Bureau of Standards.

2. Validity of Secondary StandardsA. Expression of the result:

Let us consider for example the calibration certificate of a solution of  $^{32}\text{P}$ . Let us assume it indicates:

$$3.7 \times 10^4 \text{ dps/g} \pm 1\% \text{ on 1.1.1968 at 1200 hrs.}$$

How accurate is this figure?

B. Radioactive purity:

On the face of it a standard must be pure, which means to say that the impurity content must be of the second degree at least in relation to the error indicated. In our case, the radioactive impurity content should be less than 0.1%. This is particularly not so for  $^{32}\text{P}$  which contains from 0.2 to 3%  $^{33}\text{P}$ .

If this reasoning is applied to the everyday production of radioisotopes, it will be seen that only 50, out of every 90 commercially produced radionuclides delivered, meet this specification. What should the metrologist do in such a case? Should he deliver standards only when the radioisotopes are really pure and thereby agree considerably to limit the scope of application, or should he, on the contrary, agree to deliver standards containing impurities and allow for them when expressing results? We have opted for this latter alternative, although it is spiritually not very satisfactory and even meets sometimes with insuperable difficulties such as:

Non-identification of the impurity or impurities,

Difficult determination of the impurity content,  
for example  $^{33}\text{P}$  in  $^{32}\text{P}$ .

### C. Decay Scheme:

Among the various measuring techniques, the coincidence methods are presently considered to be the most accurate. But in order to use these methods it is necessary to know all about the decay scheme. Now this is far from always being the case, since quite a number of currently used radioisotopes have very indefinite schemes. This is so for instance with  $^{133}\text{Ba}$ ,  $^{140}\text{Ba}$ ,  $^{110\text{m}}\text{Ag}$ , and  $^{170}\text{Tm}$ .

Furthermore, the supposedly well known schemes still produce surprises. Thus Mr. Spornol of the BCNM recently discovered a new scheme for  $^{60}\text{Co}$ .

For the validity of a standard to be acceptable, it is therefore necessary to study, or study again, each decay scheme and also to deliver with the standard the decay scheme utilized when the calibration was effected, in order that the user can make the necessary adjustments should there be any modifications.

### D. Radioactive Half-life:

This is a very important parameter because it conditions the validity of the standard in the course of time. Although measuring techniques have improved over the last few

years, large differences are unfortunately still noticed today where half-life values are concerned. Table 1 gives a number of half-life values already published in Vienna in 1966, but which highlights two aspects of the problem:

1.  $^{24}\text{Na}$  case - a short-lived element, relatively easy to determine, where the values extend from  $14.96 \pm 0.01$  hrs (Campion, 1957) to  $15.05 \pm 0.02$  hrs (Wyatt, Lyon, etc., 1961).
2.  $^{133}\text{Ba}$  case - a long-lived and fairly difficult element to measure, where deviations are significant since the recent values observed varied from  $10.2 \pm 0.8$  years (Wyatt, Reynolds 1963) to  $7.8 \pm 0.1$  years (LMR, 1967).

TABLE I. RADIOACTIVE HALF LIVES

Radio-nuclides	Determinations since 1956		This work	
	Minimum value	Maximum value		
$^{56}\text{Mn}$	$2.56 \pm 0.03$ h	$2.591 \pm 0.008$ h	$2.582 \pm 0.009$ h	
$^{60}\text{Co}$	$5.20 \pm 0.03$ y	$5.33 \pm 0.04$ y	$5.27 \pm 0.02$ y	
$^{131}\text{I}$	$8.054 \pm 0.010$ d	$8.067 \pm 0.007$ d	$8.073 \pm 0.008$ d	
$^{24}\text{Na}$	$14.959 \pm 0.011$ h	$15.05 \pm 0.02$ h	$15.00 \pm 0.02$ h	
$^{51}\text{Cr}$	$27.701 \pm 0.006$ d	$28.04 \pm 0.16$ d	$27.7 \pm 0.2$ d	
$^{54}\text{Mn}$	278 $\pm 5$ d	$313.5 \pm 0.7$ d	$312.2 \pm 0.9$ d	
$^{95}\text{Nb}$	$35.0 \pm 0.1$ d	$35.8 \pm 0.5$ d	$35.0 \pm 0.2$ d	
$^{133}\text{Ba}$	$7.2 \pm 0.3$ y	$10.7 \pm 0.2$ y	$7.8 \pm 0.1$ y	
$^{203}\text{Hg}$	$45.4 \pm 0.5$ d	$47.2 \pm 0.7$ d	$47.0 \pm 0.1$ d	
$^{204}\text{Tl}$	$3.56 \pm 0.05$ y	$4.11 \pm 0.13$ y	$3.85 \pm 0.04$ y	

Table 2 shows the most recent results obtained in our laboratory and it also evidences the not inconsiderable deviations, particularly for  $^{170}\text{Tm}$ .

TABLE II. RADIOACTIVE HALF LIVES

Radio-nuclides	Determinations since 1956		This work
	Minimum value	Maximum value	
$^{32}\text{P}$	14.233 $\pm$ 0.015 d	14.58 $\pm$ 0.03 d	14.268 $\pm$ 0.042 d
$^{35}\text{S}$	86.35 $\pm$ 0.17 d	88.8 $\pm$ 1 d	87.48 $\pm$ 0.40 d
$^{87}\text{Sr}^{\text{m}}$	2.83 $\pm$ 0.02 h	2.90 $\pm$ 0.08 h	2.793 $\pm$ 0.009 h
$^{99}\text{Tc}^{\text{m}}$	6.00 $\pm$ 0.10 h	6.13 $\pm$ 0.05 h	6.031 $\pm$ 0.036 h
$^{110}\text{Ag}^{\text{m}}$	250.8 $\pm$ 0.3 d	295 $\pm$ 7 d	250.38 $\pm$ 0.90 d
$^{113}\text{In}^{\text{m}}$	1.70 $\pm$ 0.03 h	1.75 $\pm$ 0.25 h	1.658 $\pm$ 0.004 h
$^{170}\text{Tm}$	125 $\pm$ 2 d	134.2 $\pm$ 0.8 d	127.10 $\pm$ 0.92 d
$^{199}\text{Au}$	3.14 $\pm$ 0.02 d		3.129 $\pm$ 0.011 d

If we also assume that the acceptable error over the half-life should be of the second degree in relation to the final error on the standard, we can establish a valid time for the standard which for example would be: 15 days for  $^{133}\text{Ba}$ , one day for  $^{32}\text{P}$ , and  $^{170}\text{Tm}$ , and three hours for  $^{24}\text{Na}$ .

#### E. Expressing the error:

This is a major philosophical issue about which much has already been written. Should a distinction be made between systematic errors and statistical errors? Should a probability of 65% - 90% or 99.7% corresponding to 1, 2 or 3  $\sigma$  be chosen? How are errors combined? By dealing with them quadratically without distinction or by

dealing with the random errors alone? All these are difficult questions to answer. Nevertheless there is an evident conclusion to be drawn: the error ranges do not tally when the various known values for the same half-lives are examined (Figures 1, 2) or when the results of an international intercomparison are summarized (Figure 3).

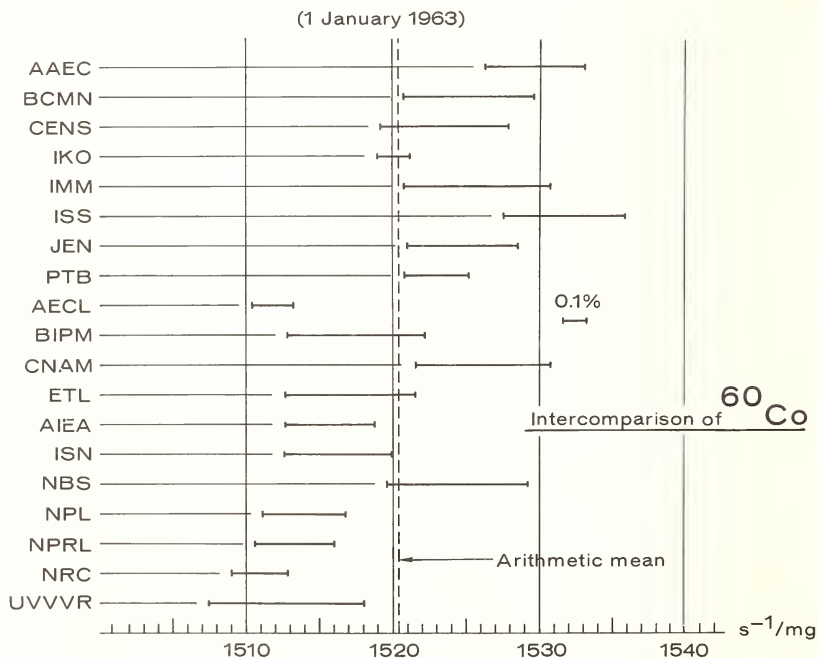


Figure 1.



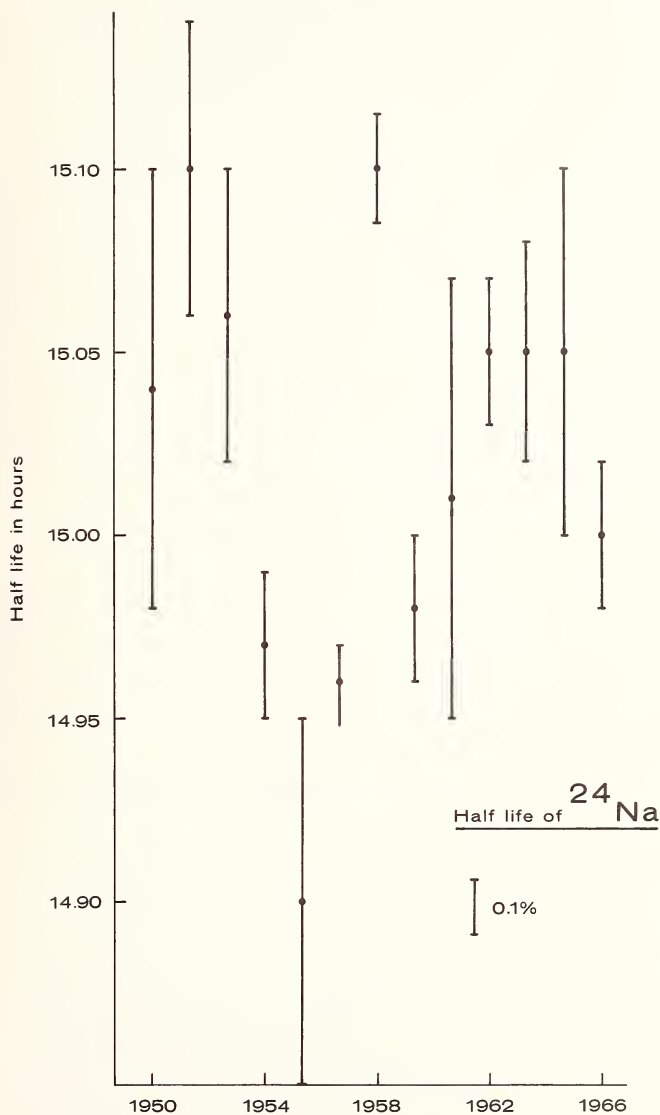


Figure 2.

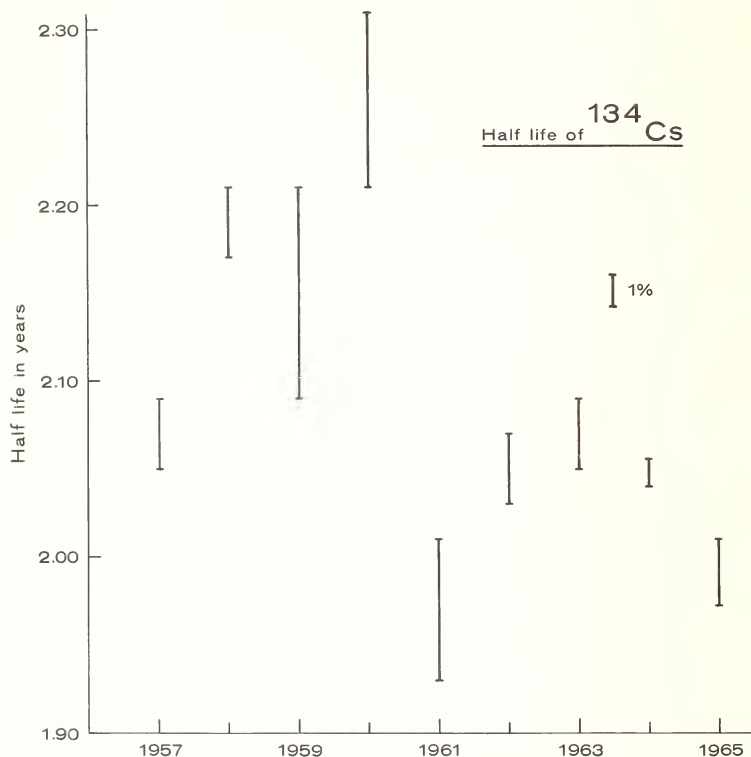


Figure 3.

One has therefore no alternative but to agree that the authors are optimistic, to say the least, and that if the same ideas were applied to radioactivity standards one would be entitled to doubt their worth. This is why a number of laboratories, including our own, has decided to deal quadratically with random errors to express the error, by adopting

a probability of 0.997 which corresponds to  $3\sigma$ , and to find the arithmetic sum of the errors known as systematic, S. These two kinds of errors are then combined arithmetically,  $3\sigma + S$ .

This philosophy of errors is not quite strict; in particular it is not correct to combine systematic errors by finding their arithmetic sum. We are, of course, aware of this but prefer to be cautious rather than optimistic where metrology is concerned.

This point of view has another drawback, needless to say, it lowers the value of the standards of the laboratory expressing the error in this way as compared to those who use for example, an "effective standard deviation" such that

$$E S D = \frac{1}{6} \quad [ (3\sigma + S) = (3\sigma + S') ] .$$

From this it will be seen that the accuracy expressed with an E S D will be three times better than that achieved by using the formula we advocate, but then these are commercial considerations which have nothing to do with metrology.

F. Validity of secondary standards:

In point of fact, from what has been stated, the validity of a secondary radioactive standard frequently depends much more on the foregoing factors than on the standard of measurement as such. This is obviously no excuse for not improving the measurements, but it does go to show that radioactive metrology research is not uniform.

### 3. The Primary Standards Problem

The question of the validity of the determinations effected in the national laboratories remains to be posed. In theory this is the job of the International Bureau of Weights and Measures (IBWM), but the task to be assumed is overwhelming and it is hardly possible to undertake more than one intercomparison every other year. Furthermore it is often difficult to put the results to use. Figure 3 which summarizes an intercomparison of  $^{60}\text{Co}$  made in 1965, illustrates the following points:

- A. A dispersion of 1.5% is observed and the arithmetical average shown on this figure is only for the record.
- B. It is impossible to quote the activity of this  $^{60}\text{Co}$  solution to 0.1% accuracy, but the most that may be said is that the most likely activity is determined within a bracket of  $\pm 0.5\%$  and this is far too imprecise.
- C. It may also be assumed that one group of laboratories using a given weighing method may work with, say greater accuracy than another group using another method. But the inter-comparison does not show this up for many errors can offset each other, and as a consequence this requires extra studies that each laboratory must undertake for itself.

This gives rise to the problem of measurement reliability and, thus, of the national primary standards, since if each

national laboratory taking part in this  $^{60}\text{Co}$  inter-comparison had calibrated - in parallel with the solution delivered by the IBWM - a solution or sources which would subsequently form its primary standards, we would now find ourselves face to face with the following situation:

1. In different countries there would be primary standards of activity which would sometimes differ by several percent.
2. With accurate transfer instruments it would be possible to obtain excellent secondary standards relative to these primary ones, which could in turn be internationally inter-compared, thereby enabling the divergencies to be elucidated.
3. It would then be possible to refer back, define the true value of the standards as inter-compared several years before, adjust the value of any erroneous national primary standard and, above all, advise the users of the secondary standards delivered by this same national laboratory as to adjustments to be made on them.

In this spirit our laboratory has undertaken the development of the primary standards of activity, having selected the following as reference radionuclides:

Solid phase :  $^{60}\text{Co}$      $^{55}\text{Fe}$      $^{241}\text{Am}$

Liquid phase :  $^3\text{H}$      $^{14}\text{C}$      $^{60}\text{Co}$      $^{90}\text{Sr}$      $^{133}\text{Ba}$      $^{241}\text{Am}$

Gas phase :  $^3\text{H}$      $^{14}\text{C}$

To conclude, we consider that the validity of radioactivity standards depends on the following:

1. The development of primary standards in each metrological laboratory.
2. Joint international action to undertake the study of fundamental constants (decay scheme, half-life etc.).
3. International agreement to standardize the statement of results and errors.

It should be finally observed that activity units must not be the sole preoccupation of nuclear metrology laboratories. This is why it seems to us also necessary to undertake the development of exposure rate and energy fluence primary standards.

W. B. Mann

Are there any questions for M. LeGallic? Dr. McNair:

A. McNair

I think it's not so much a question as a comment. I think that M. LeGallic's point about impurity is quite an important one and I think that standardizing laboratories are in danger of operating in rather a vacuum by not really being too concerned about what the users of the standard want. In some ways I think it is better to prepare a user a standard which has the same nuclidic composition as the type of material that he is wanting to standardize. A

particular case which comes to mind here, is some discussion that we had with a user of strontium-89 standards which he got from us. We produce our strontium-89 for standards work by irradiation of enriched strontium-88 because this gives a lower impurity figure and a more easily measured one because the main impurity is strontium-85. But this user, without telling us, wanted to use this standard to calibrate fission-product strontium-89 where the impurity, of course, is strontium-90. It was quite useless for this purpose, although in fact it was a much purer material than he would have had if we had used strontium-89 from fission-product material. I think that we are always running into this danger if we try to improve purity to the ultimate extreme and we may find that the resultant standards are of little use to the user.

Y. LeGallic

I agree with your point.

W. B. Mann

Thank you. In the early days when we were standardizing phosphorus-32, there was always phosphorus-33 present, but we didn't worry about this because the medical people and the hospitals got the same samples from Oak Ridge which, of course also had phosphorus-33 present.

I would like to make just two comments. Apropos the certification of standards, there is a very valuable and interesting report out, ICRU #12 by Mr. Garfinkel and Mr. Newbery of the Radiochemical Centre, which might be of interest to people.

And another comment that I have. I recall that the Russians were in the habit so I was told, I've never seen a certificate, of stamping their certificates that the validity of this certificate expires after such and such a date, and this is something perhaps that we'd want to consider, that one can cancel out a certificate after its run so many days, so many weeks, so many years.

W. B. Mann

The next paper is by Mr. J. G. V. Taylor from Chalk River, Atomic Energy of Canada Limited, and it is entitled: "A Review of Some Applications of Radioactivity Standardization Methods to the Determination of Nuclear Data".  
Mr. Taylor:

J. G. V. Taylor

Mr. Chairman and Colleagues, the title which Dr. Mann gave you is indeed what I intended to speak on, but some time after that, Dr. Mann asked if Dr. Baerg and I would say something about radioactivity standardization in Canada in general.



Radioactivity Standardization in Canada

J.G.V. Taylor

Atomic Energy of Canada Limited,  
Chalk River Nuclear Laboratories

Chalk River, Ontario, Canada

and

A.P. Baerg

National Research Council, Ottawa, Canada

In Canada, two government organizations are engaged in radioactivity standardization.

The National Research Council (NRC) in Ottawa has responsibility for fundamental physical standards, and in this respect its position is somewhat analogous to that of the National Bureau of Standards, the National Physical Laboratory, and other national standardizing laboratories. The NRC group engaged in radioactivity measurements (under A. P. Baerg) is in the X-Rays and Nuclear Radiations Section of the Applied Physics Division and is associated with other groups involved with standards of ionizing radiations (X-rays under C. Garrett and W. H. Henry, and neutrons under K. W. Geiger). L. E. Howlett, the Division's recently retired Director, was from 1964 to 1968 President of the International Committee of Weights and Measures which sponsors international collaboration in radioactivity standardization through the International Bureau of Weights and Measures

(BIPM). C. Garrett, K. W. Geiger and A. P. Baerg are members of the BIPM working group on X-ray, neutron, and radioactivity standardization, respectively and A. P. Baerg is a member of the NAS-NRC (USA) Subcommittee on the Use of Radioactivity Standards.

The other Canadian government group active in radio-nuclide standardization is at the Chalk River Nuclear Laboratories (CRNL) of Atomic Energy of Canada Limited (AECL). AECL is a Crown Corporation, that is, an establishment wholly owned by the government but set up as a company to allow more administrative and commercial flexibility than a department of the Civil Service. The position of CRNL is analogous to that of other national atomic energy laboratories such as those at Oak Ridge or Harwell. J. G. V. Taylor of the Radioisotope Standardization Section in the General Physics Branch at CRNL is also presently a member of the NAS-NRC (USA) Subcommittee on the Use of Radioactivity Standards.

Historically, the Chalk River laboratories are an offshoot of the National Research Council in Ottawa. Radium standardization began at NRC in 1930 as part of its work in radiology. Then the wartime atomic energy program resulted in the Chalk River Project which became a division of NRC. At Chalk River, absolute counting techniques were developed to meet the needs of nuclear physics and chemistry, reactor technology, and radioisotope production programs. During the years following the formation of AECL in 1952, NRC had to make up for the loss of the Chalk River group by expanding its Ottawa standardization facilities to meet its statutory responsibilities in this field. This is why, during the 1950's, it was AECL rather than NRC which collaborated with the NBS and NPL in international comparisons of radioactivity standards 1/.

Since, except for a few long-lived nuclides, it is not possible to maintain physical standards of radioactivity analogous to a standard kilogram, laboratories responsible for radioactivity standardization must concentrate on maintaining and improving apparatus and methods for making absolute measurements. In common with many other laboratories, NRC and CRNL depend on  $4\pi\beta\text{-}\gamma$  coincidence counting as the primary absolute method.

At NRC, measurements are made in response to requests and intercomparisons are carried out with other national and international laboratories. The main effort, however, is directed toward basic research on absolute methods. In recent work, the emphasis has been on theoretical and experimental studies of the optimum experimental conditions and methods of data analysis for the determination of efficiency functions 2,3,4/. These empirical functions allow for the influence of complex decay schemes and other effects and are determined experimentally for each measurement. At NRC, this is done by varying the electronic discrimination level in the  $4\pi\beta$  channel. This method requires a  $4\pi$  proportional counter operated at high pressure. The counter is designed for flow operation with automatic gas-density regulation 3/.

Figure 1 is a photograph of one of the NRC  $4\pi\beta\text{-}\gamma$  coincidence systems incorporating a pressurized  $4\pi$  counter. The PDP-8S on-line computer controls the operation of the system to collect efficiency function data. When the efficiency functions are known in advance, the disintegration rates are given immediately by the computer. More elaborate data-fitting computations are handled by an IBM-360 computer via a teletype link located in the laboratory.

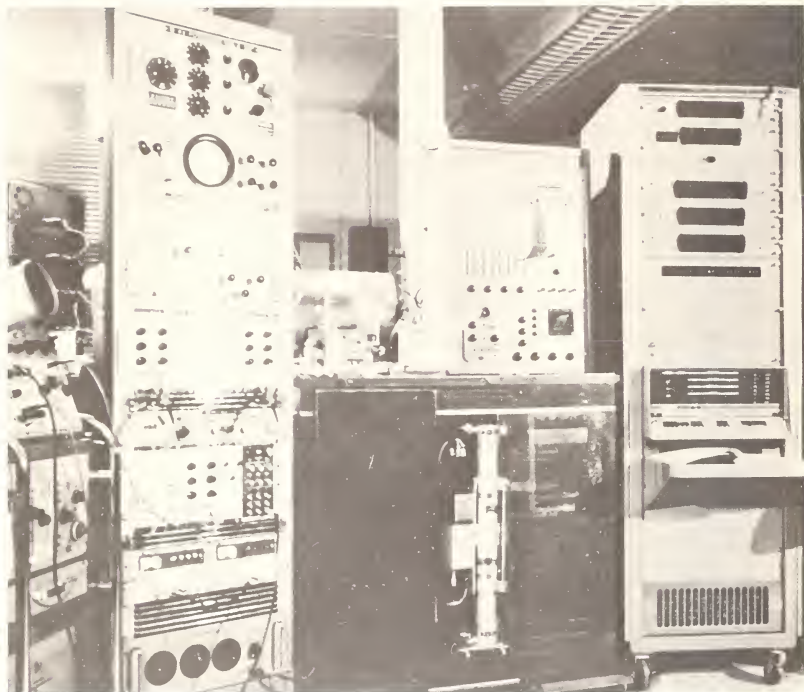


Figure 1. NRC  $4\pi\beta\text{-}\gamma$  coincidence apparatus. The detectors can be seen through the open door of the castle in the center. The computer-based data acquisition and control equipment is on the right; the rest of the electronics and the gas density regulating system are on the left.

Current research at NRC includes studies of resolution and gain stability in  $4\pi$  proportional counters, coincidence measurements of electron-capture decay, and decay-scheme investigations using absolute activity measurements and high-resolution electron and photon spectroscopy. Recently a wall-less multiwire,  $4\pi$  proportional counter for electron-capture studies has been set up by R. J. Adams.

At CRNL, the general picture is similar. Standardizations are made as requested and the group participates in international and other intercomparisons of standards. The research emphasis is on improving the techniques and apparatus for radioactivity standardization and on using absolute counting methods to determine decay parameters and other nuclear data. 5,6,7/ P. J. Champion's original Chalk River  $4\pi\beta$ - $\gamma$  coincidence apparatus 8/ recently has been modified to include an automatic sample changer. The very fragile source mounts did not permit the use of any commercially available changer. In Figure 2,

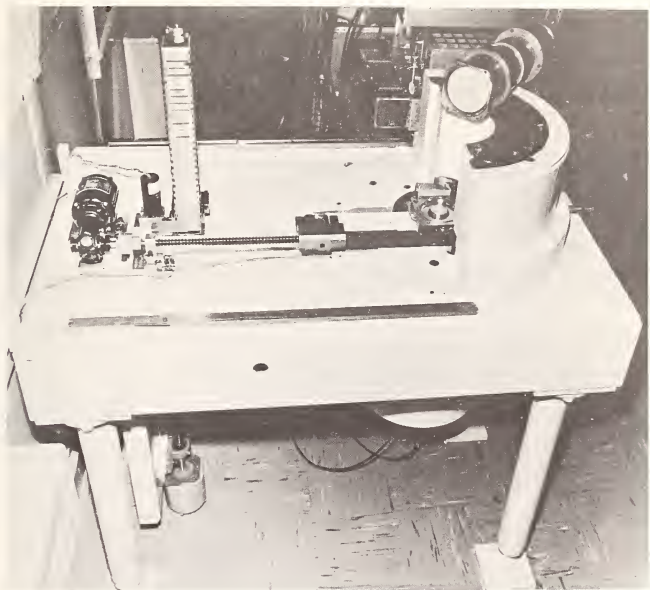


Figure 2. CRNL  $4\pi\beta$ - $\gamma$  coincidence system partly dismantled to show the automatic sample-changer details. The digital stepping motor under the table to the left positions the 36-sample vertical magazine stack to select the sample. A sample in the sample carrier (center) can be seen half way between the stack and the  $4\pi\beta$  counter. The upper NaI(Tl)  $\gamma$ -ray detector has been raised and laid across the only piece of the upper lead shielding not removed for this picture.

much of the shielding has been dismantled and the upper  $\gamma$ -ray detector moved to show the  $4\pi\beta$ -counter, the sample insertion and withdrawal mechanism with a sample halfway between the counter and the magazine stack, and the sample-selecting mechanism which drives the 36-sample stack vertically. Figure 3 is a general view of the associated electronics. The output data are stored on punched paper type which is sent to the computation center. The system was intended for use with a PDP-8 computer but the interface for this is not yet available. Other equipment used at CRNL includes a precision high-pressure  $4\pi\beta$  ion-chamber system for  $\gamma$ -ray measurements, a large pressurized  $4\pi$  x-ray proportional counter for electron-capture measurements detectors for x-ray - x-ray coincidence

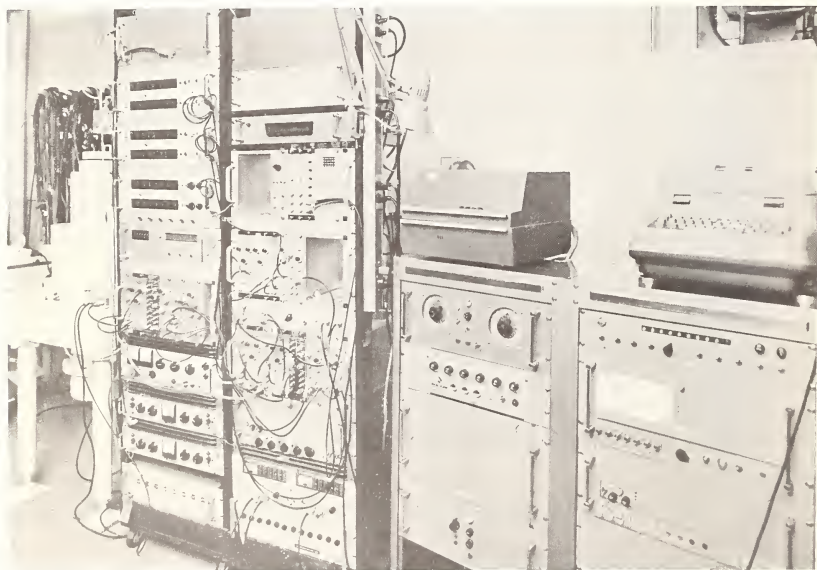


Figure 3.

The electronics for the CRNL  $4\pi\beta$ - $\gamma$  apparatus. The temporary sample-changer control and data readout system is on the right. The assembled castle is partly visible on the left.



counting, and sets of compensated gas counters for gas-phase counting of tritium,  $^{14}\text{C}$ ,  $^{85}\text{Kr}$ , etc.

Radioactivity standards are not issued on a regular schedule by either government laboratory because this would not be justified by the present Canadian demand. Both organizations attempt to fill any requests they receive, especially if the requirements cannot be met by standards available from the IAEA, NBS, RCC or other commercial suppliers.

Turning to universities, L. Yaffe, well known for his studies of  $4\pi\beta$  counting at McGill in the mid-1950's, still maintains absolute-counting facilities but is no longer active in standardization research. Nor is his collaborator in that work, B. D. Pate, who is now at Simon Fraser University. J. C. Roy at l Université Laval recently has been studying source-preparation techniques for  $4\pi\beta$  counting, especially electrodeposition methods 9/.

In closing we would like to draw attention to the major contribution to radioactivity standardization represented by the alumni of the Chalk River Project dating from both the NRC and AECL eras. P. J. Campion, who heads the Radiation Science Division at the National Physical Laboratory in England, spent six years at Chalk River where he became well known for his work establishing  $4\pi\beta\text{-}\gamma$  coincidence counting as the primary method in radionuclide standardization. A. P. Baerg, presently at NRC, spent a considerable part of his seven years at Chalk River doing absolute counting in connection with fission-yield and cross-section measurements. While at Chalk River, L. Yaffe was a prominent figure in the pre- $4\pi$ -counter era of absolute measurements; his detailed studies of  $4\pi\beta$  counting at McGill followed up the pioneer investigations of this method at Chalk River by R. C. Hawkins and W. F.

Merritt. J. C. Roy, now of Laval, also spent seven years at Chalk River before going to the BIPM in Paris for two years. Finally, the Chief of the NBS Radioactivity Section and chairman of this session, W. B. Mann, was at Chalk River for three years in the late 1940's when, among other achievements, he developed his first set of compensated internal gas counters for absolute determinations of  $^{14}\text{C}$  to measure its half life.

W. B. Mann

Are there any questions for Mr. Taylor?

Anon

Such a good talk, there are no questions.

W. B. Mann

Thank you, Mr. Taylor. I'm honored to be an alumnus of so distinguished a laboratory.

One of the very exciting laboratories I've visited in recent years is the Boris Kidric Institute in Yugoslavia. Dr. Bek-Uzarov has very kindly come at short notice to talk about the work in the Radioactivity Group at the Boris Kidric Institute. Dr. Bek-Uzarov.



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Metrology of Radioactive Isotopes at the Boris Kidric  
Institute of Nuclear Sciences

by

Dj.N. Bek-Uzarov

Vinca-Beograd, Yuogslavia

A survey of the activities of the department of Radioactive Isotope Metrology of the Boris Kidric Institute of Nuclear Sciences at Vinca is given. Some new problems with which this Department is dealing in developing counters used for absolute measurement of the number of decays are presented. Work on the estimation of air-equivalent materials for the production of air-equivalent cavity ionization chambers is also surveyed.

The first problems of radioactive-isotope measurement were encountered with the needs for neutron-flux measurements when the reactor of the Boris Kidric Institute and, later, the Hot Laboratory were put into operation. In 1956, a group was formed whose task was to develop methods for absolute measurement of radioactive isotopes. Later, when the Hot Laboratory began normal work it was necessary to extend the spectrum of radioisotopes to a large number of commonly used ones which are daily produced and measured.

1. Basic Activities of the Department of Isotope Metrology
  - A. The Department deals with concepts and development of new methods of absolute and relative measurement of radioactive isotopes.
  - B. It takes part in investigations of problems of other Departments concerned with isotope measurement.
  - C. It produces radioactive standards for the entire country.
  - D. It takes part in international intercomparisons of radioactive isotopes organized by the Bureau International des Poids et Mesures, thus representing Yugoslavia in this field.
  - E. It produces and designs instruments for precise measurement and analyses of radioactive isotopes.
  - F. It cooperates with research centres and industrial laboratories in the country.

The Department is located in a separate building to protect the instruments from contamination by radioactive isotopes and to provide satisfactory electric power supply, gas (compressed air, noble gases) and water supply. The gas installations, compressed air, methane and fuel gas, are independent of the installations of the whole Institute. This supplies maximum safety in case the central installation system of the Institute fails.

The Department avails itself of the following basic instruments for absolute measurement of radioactive isotopes; beta-gamma coincidence equipment (with  $4\pi$  proportional counter and solid-angle method), two  $4\pi$  counters, apparatus for internal gas counting and apparatus for measurement of exposure gamma radiation dose rate. The relative measurements are made with:  $2\pi$  and  $4\pi$  proportional counters, a  $2\pi$  alpha ionization chamber and a  $4\pi$  integral ionization chamber for the measurement of the beta-gamma radioactive isotopes or bremsstrahlung radiation of pure beta emitters (Figures 1-7).

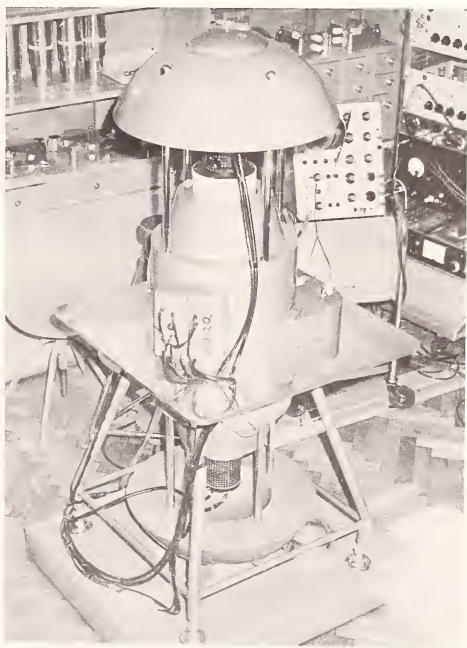


Figure 1.  $4\pi\beta\text{-}\gamma$  coincidence apparatus. Two scintillation counters with NaI(Tl) crystals, 100 mm in diameter and 60 mm in height, are visible. To the right side there is a door behind which a  $4\pi$  proportional counter is positioned.

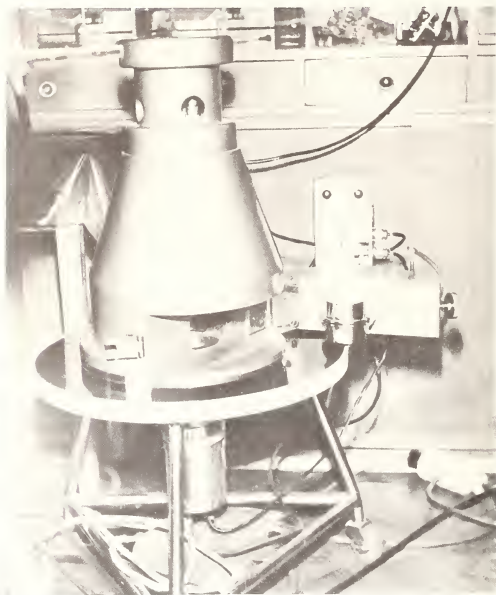


Figure 2.  $4\pi\beta\text{-}\gamma$  coincidence small apparatus. A flow counter is mounted on the door. When the door is closed the counter stands in the space between two NaI(Tl) crystals.

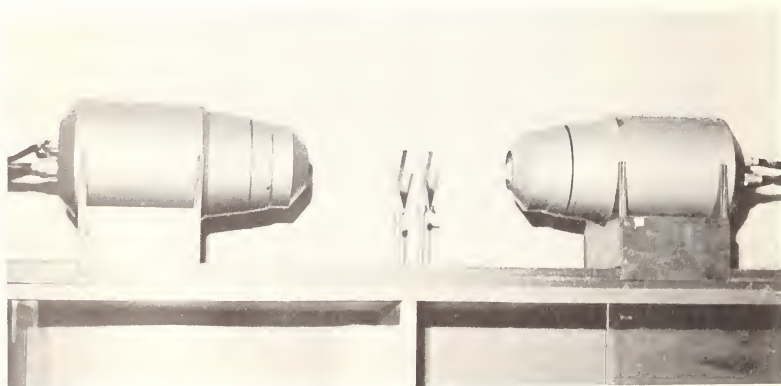


Figure 3. Solid-angle-coincidence apparatus.

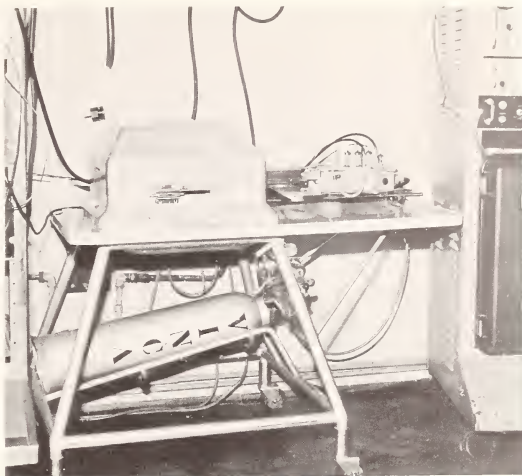


Figure 4.  $4\pi$  flow-proportional counter with a slide and a preamplifier drawn out from the lead shielding.

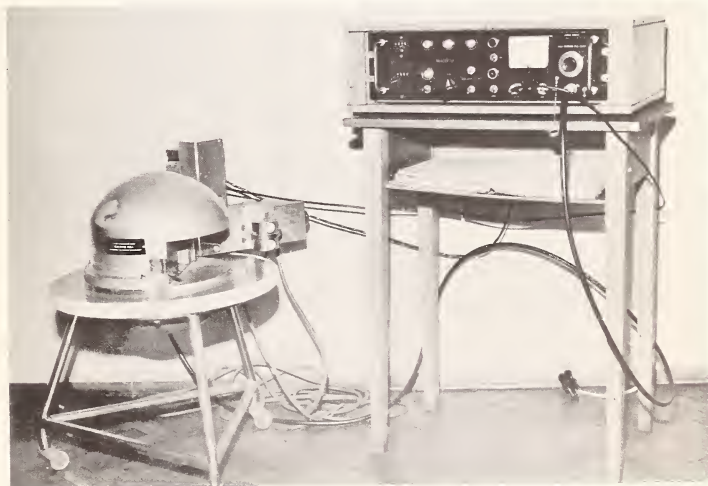


Figure 5.  $4\pi$  counter for sources of small diameter.



Figure 6.  $2\pi$  alpha-particle ionization chamber.



Figure 7. Arrangement for  $4\pi$  integral ionization chamber for relative measurements.



1) Development methods of measurement

- Efficiency of the liquid-scintillation detector.

The efficiency as a function of energy and geometry, of the  $4\pi$  liquid-scintillation counter, has been investigated.

The mean beta-particle energy absorbed in the scintillation solution, required for the production of one photoelectron on the photocathode is defined as the energy threshold for detection. The energy threshold was determined from the difference between the positions of the maximum tritium energy in a system of two electron-multiplier phototubes coupled in coincidence with a single phototube.

The dependence of the geometry factors was investigated with spherical glass flasks filled with a liquid scintillator. These flasks have different diameters and the solution is of the same "specific activity relative to the scintillator", i.e. of the same number of disintegrations per gram of scintillation solution 1/.

- Coincidence method of measurement.

In developing the coincidence method of measurement it was concluded that two types of counters were required: a real proportional and a nonproportional counter (which is called semiproportional or H-counter) (Figures 8 and 9). Semiproportional counters

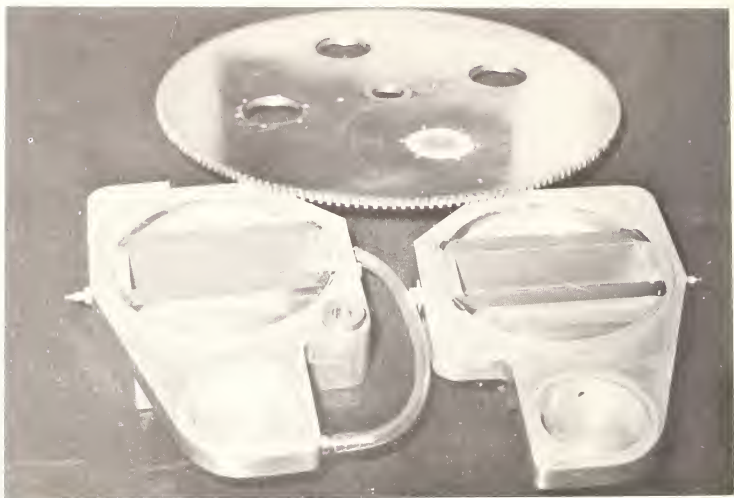


Figure 8.  $4\pi$  flow counter. The inside of the counter and a preflushing chamber with a disc sample chamber are seen.

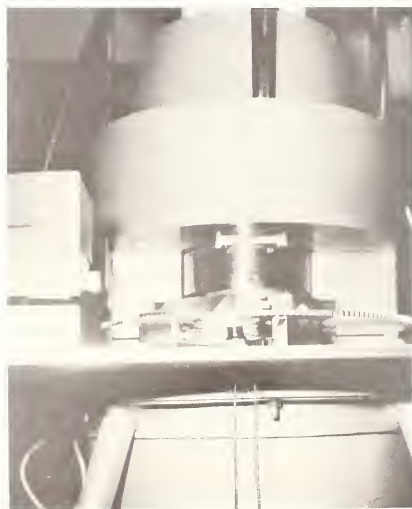


Figure 9.  $4\pi$  flow counter mounted in a coincidence apparatus.

are flow counters which are non-proportional because a semiconductor diode is put in the anode circuits instead of the operating resistance. By employing the diode and a nonoverloading amplifier, one obtains a counter which is very much like a GM counter except for its counting rate, which is many times faster.

Figure 10 shows results of the comparison of pulse-height-spectral distribution. In the upper part of the graph there are integral spectra obtained after pulse amplification so that their maxima do not exceed 80 V. It is apparent that the slope of the integral spectrum for the H-counter is almost a straight line. The magnitude of the slope is compared using the conventional formula:

$$D_s = \frac{(N_2 - N_1) \cdot 100}{\frac{N_1 + N_2}{2} (V_2 - V_1)} \quad [\% \cdot V^{-1}], \quad \dots (1)$$

where  $N_1$  denotes the counting rate at the beginning of the integral curve (figure 10) where the magnitude of the pulse amplitude is  $V_1$ ;  $N_2$  denotes the counting rate at the end of the rectilinear part of the curve where the pulse amplitude is  $V_2$ .

Because of the different counting rates all curves were normalized. In the lower part of the graph, besides the differential spectra, forms of analyzed pulses with time parameters

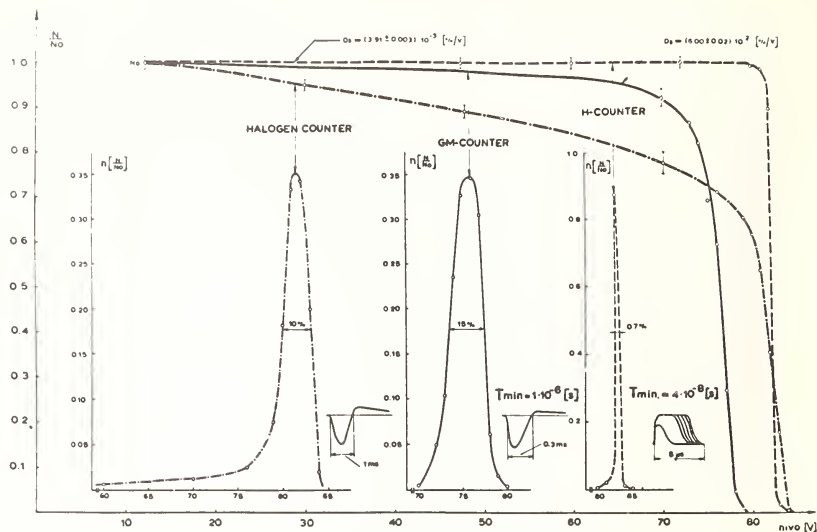


Figure 10. Differential and integral pulse-height spectra taken with halogen, GM and H-counters.

are shown.  $T_{\min}$  denotes the minimum rise time which could be estimated and beside the differential spectrum, estimation of the pulse time is given. For the first two counters the pulse form at the output of the amplifier is shown. The use of the limiter in the nonoverloading amplifier prevented cutoff in any circuit.

To estimate the relative width of differential spectra  $R_S$ , the following expression has been adopted:

$$R_S = \frac{\Delta V}{V_P} \frac{N_1}{N_P} 100 [\%], \quad \dots (2)$$

where  $V_p$  denotes the pulse amplitude in the peak of the spectrum, while  $\Delta V$  denotes the peak width at half the height (in voltage);  $N_p$  denotes the normalized counting rate in the peak and  $N_1$  the counting rate at the beginning of the integral curve.

Figure 11 shows the characteristics of plateaus (I, II, III) when resistors and diode are employed on the counter. By using a higher amplification factor of the amplifier we obtain a plateau at lower voltages which is shown by curve IV. Curve V shows a plateau with an inversely polarized diode (in which case the diode is used as the current limiter). The plateaus for pure alpha-particle counting are about 1500 V below beta (or alpha + beta)

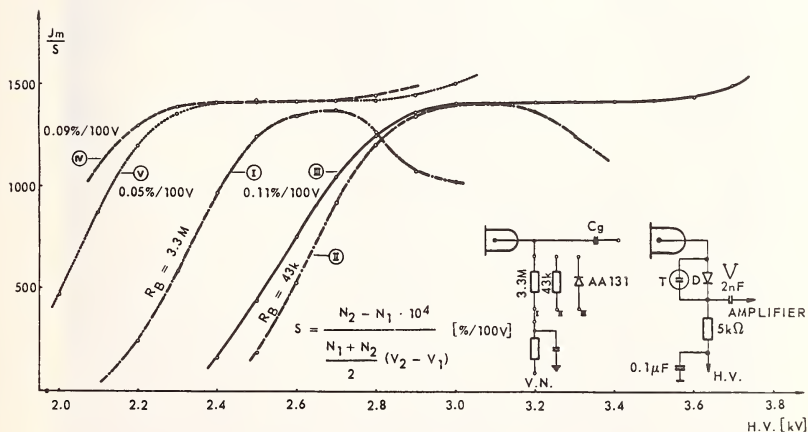


Figure 11. Different plateaus obtained by using resistors and semiconductor diodes in anode circuit of methane flow counter.

counting plateaus. The beta (or beta + alpha) plateaus shown in figure 11 are obtained using cobalt-60 sources.

All elements of the input circuit of the counter are immersed in oil or epoxyde. The diode is protected from too high an inverse voltage by using small gaseous 60-V discharge tubes denoted by T.

To measure electron-capturing nuclides, a proportional counter filled with a mixture of argon (90%) and methane (10%) with constant pressures, up to 10 atmospheres, is used (figure 12). The sides of the counter facing



Figure 12. Proportional counter for measuring the number of decays of electron-capture nuclei.

towards NaI(Tl) crystals can be replaced by thinner plates if absorption of radiation is to be reduced when making coincidence measurements.

- Air equivalent ionization chamber for measuring the exposure gamma radiation dose rate.

Data are available 2,3/ which assist in the choice of materials for the construction of air-equivalent cavity chambers. In our case, too, a cavity-type ionization chamber has been made of air-equivalent material used for measuring gamma radiation in roentgen units. The air-equivalent material has also been estimated on the assumption that mass-absorption coefficients for the photoelectric, Compton effect and pair production are equal to the product of two functions: the function depending on the energy of gamma radiation and the function depending on the atomic number of the element 4/.

If  $\mu_f$ ,  $\mu_c$  and  $\mu_p$  denotes the mass-absorption coefficients for photoelectric, Compton and pair-productio effects respectively, then the following three relations can be written:

$$\sum_{i=1}^n p_i (\mu_f)_i = (\mu_f)_{\text{air}}, \quad \dots (3)$$

$$\sum_{i=1}^n p_i (\mu_c)_i = (\mu_c)_{\text{air}}, \quad \dots (4)$$

$$\sum_{i=1}^n p_i (\mu_p)_i = (\mu_p)_{\text{air}}, \quad \dots (5)$$

where the air-equivalent material contains the  $i$ -th element whose percentage content in the whole material is  $p_i$ . This system also involves an equation that defines the percentage composition of each component of the material of which the air-equivalent mixture is made, i.e., the following equation can be written:

$$\sum_{i=1}^n p_i = 1. \quad \dots (6)$$

Hence, the following approximation can be made, i.e. relations (3), (4) and (5) are assumed to be:

$$\mu_f = (Z^{4.1}/A) f(E),$$

$$\mu_c = (Z/A) C(E),$$

$$\mu_p = (Z^2/A) P(E),$$



where  $Z$  and  $A$  denote the atomic and mass numbers of the element concerned, while  $f(E), C(E), P(E)$  denote the functions of energy of the above phenomena; for estimating the material, it is not necessary to know the form of functions.

Using the above approximations and knowing the percentage composition, by weight, of air (76.5% nitrogen, 23.2% oxygen and 1.3% argon), the system of equations (3), (4), (5) and (6) can be reduced to the following system:

$$\sum_{i=1}^n a_{ji} p_i = b_j, \quad (j = 1, 2, 3, 4) \quad \dots (7)$$

where the coefficients have the following values:

$$a_{1i} = \frac{Z_i^2}{A_i}; \quad b_1 = 0.755 \frac{Z_N^{4.1}}{A_N} + 0.232 \frac{Z_O^{4.1}}{A_O} + 0.013 \frac{Z_A^{4.1}}{A_A} = 233.7 \quad \dots (8)$$

$$a_{2i} = \frac{Z_i}{A_i}; \quad b_2 = 0.755 \frac{Z_N}{A_N} + 0.232 \frac{Z_O}{A_O} + 0.013 \frac{Z_A}{A_A} = 0.499 \quad \dots (9)$$

$$a_{3i} = \frac{Z_i^2}{A_i}; \quad b_3 = 0.755 \frac{Z_N^2}{A_N} + 0.232 \frac{Z_O^2}{A_O} + 0.013 \frac{Z_A^2}{A_A} = 3.65 \quad \dots (10)$$

$$a_{4i} = 1 ; b_4 = 1 \quad \dots (11)$$

The system of equations (8), (9), (10) and (11) is not equivalent to the system of equations (3), (4), (5) and (6). However, the former represents the approximation to the latter, which depends on the use of the approximation formulae for mass coefficients of energy absorption.

Since the estimation has been made with certain approximations 5/ it is necessary to check the equivalence of the calculated mixtures. By calculating the relation  $\lambda$  which is defined as:

$$\lambda = (\mu_a)_{AEM} / (\mu_a)_A$$

(where AEM denotes the air-equivalent material and A denotes air) for different gamma-radiation energies, and by calculating the stopping powers of air and air-equivalent material

$$S = \frac{S_A}{S_{AEM}} \quad \text{for different electron energies,}$$

we make a definite check of the mixture of elements.

Figures 13 and 14 give the dependence of  $\lambda$  on gamma-radiation energies for the two estimated mixtures. The figures also show the dependence of  $\lambda$  and  $S$  as a function of

the gamma-radiation energy and beta-particle energy respectively. The estimation was made using the tables cited by Yoyet et al 2/ and Nelius 6/.

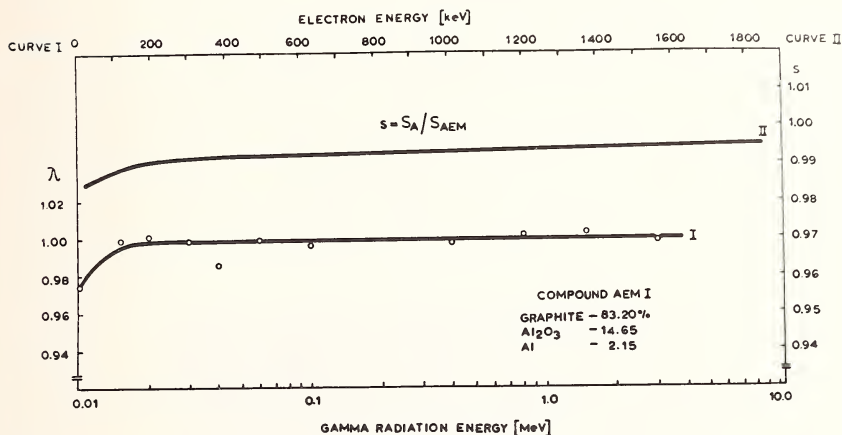


Figure 13.  $\lambda$  and S-function for AEM I mixture.

## 2) Research work in cooperation with other Departments

In cooperation with the Medical Department and the Health Physics Department, our Department has constructed an apparatus for whole-body radioactivity measurement (whole-body monitor). The equipment was made of instruments and materials produced in our country except for the crystal and steel used for the cabin. The equipment is so constructed that it can be used both for total-body

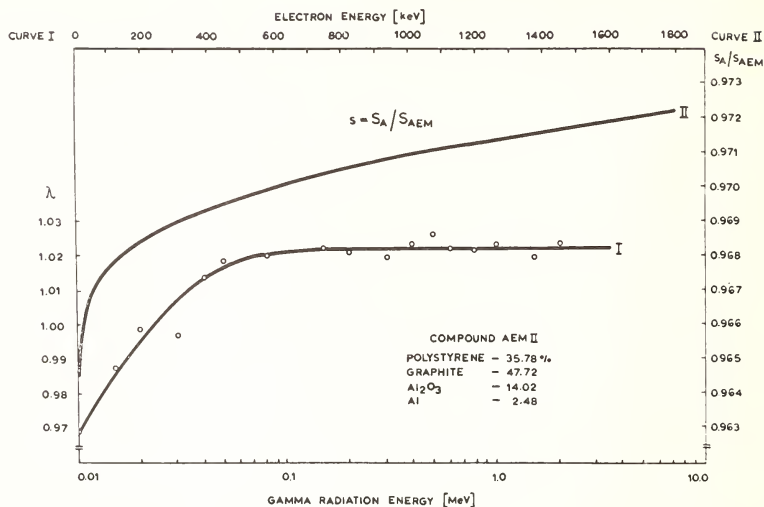


Figure 14.  $\lambda$  and S-function for AEM II mixture.

measurement and, using the scanning system, for detection of internal contamination. The construction of the cabin and the equipment for position indication has interesting original technical solutions and applications.

The production of the radioactivity standard of the human body is one of the difficult problems of radioisotope metrology.

The problem of measurement and indication of atmospheric radioactivity in nuclear power stations has also been solved in our Department by the construction of ionization chambers which measure the air ejected from the reactor.

3) Production of radioactive reference sources

As our Department has the best standard in our country it naturally undertakes the production of radioactive standards for all customers.

Solid standards ready for measurement are put in water-proof cases in which a drying agent is put to keep the series of sources always dry and ready for measurement with necessary reproducibility.

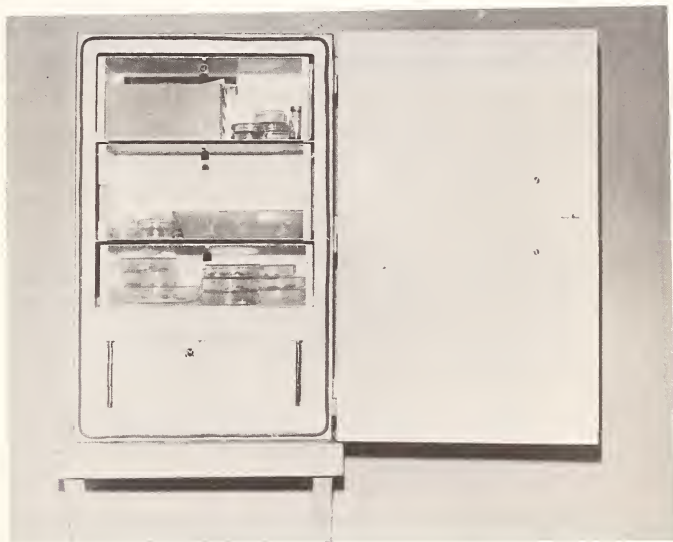


Figure 15. Case for preserving dry solid sources.

Comparisons of the radioactive sources of consumers in our country have been made twice. Our Department distributes the radioactive solutions among the interested laboratories and then collects their results of measurements. In our country we repeated what the Bureau International in Sévres had done on an international scale.

4) Participation in intercomparisons of radioactive solutions

Our Department has participated in intercomparisons ever since 1958/1959 when first organized by the International Atomic Energy Agency and later by the Bureau International des Poids et Mesures. It need not be pointed out what the usefulness of this work is. However, the burden of the work predicted in the field of intercomparison is ever growing. On the one hand this is positive, but on the participation of small laboratories in the measurements in which many people and instruments and much involvement and time are required, is more and more difficult.

5) Applied instruments for precise measurements

The results of almost every intercomparison are many new technical improvements, new constructions, some new solutions, which are used in the construction of small systems or individual instruments for different consumers in the country (Figure 16).

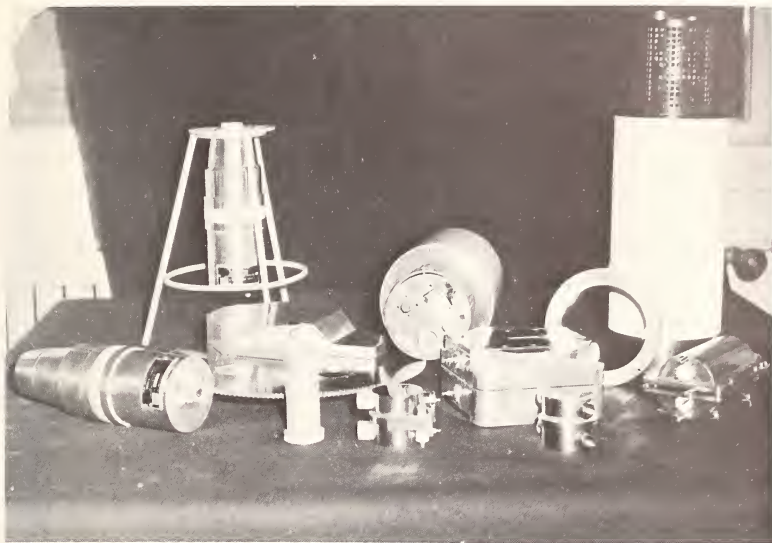


Figure 16. Some proportional and scintillation counters of home design and construction.

It very often happens that the instruments constructed and used in our Department are given to the consumers (hospitals or industrial laboratories) and then our Department designs for its own use, new, better and more perfect equipment. This is satisfactory both for our Department and the consumers: the consumers get the most up-to-date instruments and our Department gets the best ones.

Our Department designs scintillation detectors, proportional counters and ionization chambers (for alpha-and gamma-ray measurement). Naturally, the number of these instruments is not

large, only a few of them are made per year. This activity is considered fairly useful because the technical staff is always in a constant working condition.

#### 6) Cooperation with research centres

As the problems of metrology are often investigated in different laboratories, our Department most often appears as the place where both foreign and home students get preliminary training. A number of experts presently working in our Institute, or in some other institutions, have passed through this preliminary phase of training in our laboratories for six months up to one year.

There are also some regular exercises included in the programme of the University of Beograd which are carried out in our Institute.

Cooperation with Faculties is of mutual benefit. Our laboratories get young personnel while the Faculties are provided with practical work of highest quality in our country.

Physicists and engineers trained to work in the Health Physics or Medical Department, as a rule, first pass through our Laboratory.



There have been several projects concerned with the application of radioisotopes in industry in which our Department took part. Unfortunately we must point out that in this field the possibilities have not been exhausted and, so far, we have not achieved all the expected results.

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W. B. Mann

Are there any questions for Dr. Bek-Uzarov?

S. B. Garfinkel

You mentioned that you were able to reduce the dead time of a proportional counter by a factor of 20?

Dj. N. Bek-Uzarov

Yes, up to 20.

S. B. Garfinkel

How were you able to do this?

Dj. N. Bek-Uzarov

But not just the dead time. We have replaced the proportional-counter load resistor with a semi-conductor diode. As this has a non-linear voltage-current characteristic, the larger counter pulses are reduced in amplitude, so that there are many fewer pulses large enough to saturate the amplifier.

W. B. Mann

Any more questions? M. LeGallic:

Y. LeGallic

When you use your automatic sample changer what's the contribution of the sources in storage to the background?

Dj. N. Bek-Uzarov

In the case of cobalt-60 we measure only 2 sources but in other cases the contribution may be 5 to 10%.

W. B. Mann

Thank you very much, Dr. Bek-Uzarov. Finally in this program it is a great pleasure to introduce my friend and colleague Mr. Garfinkel to talk about our work at the National Bureau of Standards.



Recent Activities of the NBS Radioactivity Section with  
Special Reference to the Standardization of Thallium-208

by

S. B. Garfinkel and J. M. R. Hutchinson

National Bureau of Standards

Washington, D.C., U.S.A.

The Radioactivity Section of NBS has the mission of developing, preparing and maintaining radioactivity standards for the United States. We also distribute a majority of these standards and provide a calibration service covering almost all the radionuclides with which we have worked, particularly those we no longer distribute. This calibration service is available on a cost basis to individuals, private companies, government agencies, and research laboratories, in the United States. Some of the standards have been dropped from distribution either due to lack of demand or because they are available from commercial distributors who have demonstrated their willingness and competence to maintain their calibration facilities in good enough order to meet the requirements and demands of the public. Table 1 is a list of the radionuclides which we are distributing. Some of the radionuclides which are marked by an asterisk are not always available, either due to the short half-life or lack of customer demand. A great many techniques are used in arriving at standardized sources of these various nuclides, such as  $4\pi\beta$ ,  $2\pi\alpha$ , and  $1\pi\alpha$  counting;  $4\pi\beta$ - $\gamma$ ,  $2\pi\beta$ - $\gamma$ ,  $4\pi$ x-ray- $\gamma$ ,  $\gamma$ - $\gamma$ , and x-ray

Table I

<u>Radioactivity Standards</u>		
<u><math>\alpha</math> SOURCES:</u>	<u><math>\beta</math> GAS STANDARD:</u>	<u>RADIUM SOLUTIONS:</u>
Polonium-210	Krypton-85	For Radon Analysis: $10^{-8}$ gram
Americium-241		$10^{-9}$ gram
	<u><math>\gamma</math>-RAY POINT SOURCES (Energy in MeV):</u>	$10^{-11}$ gram
<u><math>\beta</math>, <math>\gamma</math>, EC SOLUTION SOURCES:</u>	Sodium-22 (0.511, 1.274)	Blank
Hydrogen-3	Manganese-54 (0.835)	
(water, 2 intensities;	Cobalt-60 (1.173, 1.332)	For $\gamma$ -ray Measurements:
toluene)	Zinc-65 (1.114)	10 intensities from 0.1 to 100 $\mu$ g
Carbon-14	*Yttrium-88 (1.836)	<u>UNDER DEVELOPMENT:</u>
(water, benzoic acid in	Niobium-94 (0.702, 0.871)	Nickel-63 $\beta$ -ray solution standard
toluene, hexadecane)	*Niobium-95 (0.765)	Krypton-85 $\gamma$ -ray gas standard
Sodium-22	Cadmium-109 (0.088)	Plutonium-238 $\alpha$ -source
Chlorine-36	Cesium-137 (0.662)	
Iron-55	Cerium-139 (0.166)	<u>DISCONTINUED STANDARDS:</u>
*Niobium-95	*Mercury-203 (0.279)	Scandium-46
Tin-113-Indium-113m -	Thorium-228-Thallium-208 (2.615)	Strontium-85
*Iodine-125		Strontium-89
*Cerium-144		
Cerium-Praseodymium-144	Contemporary Standard	
Promethium-147	for Carbon-14 Dating	
*Mercury-197	Laboratories:	
*Mercury-203	Oxalic Acid	

\*Not always available, due to combination of half-life and customer demand.

- x-ray coincidence counting; compensated internal gas counting; micro-calorimetry; and total-absorption x-ray counting. In addition we use the " $4\pi\gamma$ " ionization chamber and 3" x 3"-sodium-iodide crystals for relative calibrations, these detectors having been calibrated in terms of other radionuclides which had been standardized by direct counting techniques. We use the " $4\pi\gamma$ " chamber and sodium-iodide crystals to calibrate such radionuclides as tin 113 - indium 113m or cesium-137. These nuclides, in particular, are not amenable to coincidence counting as a means of determining the gamma-ray emission rates of a given source.

One of the standards, which is currently under development, is nickel-63. We have measured two samples of nickel-63 in our microcalorimeter <sup>(1)</sup>. To give you an idea of what is involved here: the two nickel-63 samples have an activity of  $\sim 1\text{-}1/3$  curies each, which corresponds to a heat output of 133 microwatts. We have not yet completed calculations on the total activity.

In the case of krypton-85, we have in the past issued only  $\beta$ -ray gas standards. These consist of sealed ampoules containing  $\sim 1$  microcurie of krypton-85. In order to use these, it is necessary to transfer the gas from the ampoule into the measuring system and to determine the quantity of the gas and its activity by means of  $\beta$ -particle counting or ionization-current measurements. These samples are calibrated in units of activity per mole of krypton. Because of the low abundance ( $\sim 0.4\%$ ) of the  $\gamma$  ray in krypton-85, these  $\beta$ -particle gas standards do not permit of accurate  $\gamma$ -ray counting. We have prepared  $\sim 100$  ampoules of krypton-85  $\gamma$ -ray standards which are about three orders of magnitude stronger in activity, ( $\sim 1$  millicurie each). These will be individually calibrated as to total activity, and will be able to be used without opening the ampoule. This sort of sample is of interest to people who work with very strong krypton sources such as in fuel-burn-up studies.

A recently acquired facility of the Radioactivity Section and the National Bureau of Standards is the isotope separator. Although not obtained only for use in radioactivity standards, we hope to exploit the capabilities of this instrument. One case in point is the production of niobium-94  $\gamma$ -ray standards. This is a very useful radionuclide having a half-life of some 20,000 years, and a decay scheme very similar to that of cobalt-60 except for the  $\beta$ - and  $\gamma$ -ray energies. Niobium-94 is produced by irradiation of stable niobium-93 in

a reactor and this, of course, requires a long irradiation, of the order of 2 years. With a neutron flux of about  $10^{14} \text{ s}^{-1} \text{ cm}^{-2}$ , one ends up with activities of the order of a millicurie per gram of niobium but, in addition to niobium-94, one obtains niobium-93m and niobium-95. As the latter has a 35-day half-life, one can wait a year or two for it to decay, but the longer-lived niobium-93m, having a decay energy of 30 kilovolts, would still preclude the use of  $4\pi\beta\text{-}\gamma$  coincidence counting of the niobium-94. In the past we have counted the niobium-94 by  $\gamma\text{-}\gamma$  coincidence counting. This can be done quite easily and allows one to discriminate against the niobium-93m. However, we hope to obtain some more niobium-94 and niobium-93m material and separate these in the isotope separator (provided that the yields are sufficient) and thus get two useful materials out of this: first of all, enough carrier-free niobium-94 for a laboratory working standard which we could calibrate by an independent method, ( $4\pi\beta\text{-}\gamma$  coincidence counting), and also some niobium-93m which, depending on the measurement, may possibly provide a rather nice, low-energy standard of  $\sim 10$ -year half-life. (This half-life is not well known.)

A  $\gamma$ -ray standard which has been developed recently and which has been in demand for some time is that of 1.9-year thorium-228. The  $\gamma$  ray of interest, 2.614-MeV, arises in the decay of the last radioactive member of the decay chain (3.1-minute thallium-208, Figure 1). Since an intermediate member of the chain is a gas, radon-220, it is important for the standard sources to be made in such a manner that the radon is contained within the source. The standards, for distribution, were in the form of thorium-228, sandwiched between two layers of 0.0019-cm-thick gold foil, which were in turn sandwiched between double layers of 0.036-cm-thick polyurethane film tape. Tests showed that this configuration allowed no radon to escape. Gold foil was used to protect the plastic tape from damage which might arise as a result of  $\alpha$ -particle irradiation. These



Radionuclide	Half-life	$\alpha$ -particle energies MeV	$\gamma$ -ray energies MeV
Thorium-228 (Radiothorium) ↓	1.91 y	$\sim 5.2$ — 1% 5.338 — 28% 5.421 — 71%	0.084 — $\sim 2\%$ others — very weak
Radium-224 (Thorium X) ↓	3.6 d	5.445 — 4.9% 5.681 — 95%	0.241 — 3.7% others — very weak
Radon-220 (Thoron) ↓	54 s	6.28 — $\sim 100\%$	—
Polonium-216 (Thorium A) ↓	0.158 s	6.775 — $\sim 100\%$	— ----- —
0.04% 99.96% ↓ Astatine-216 ↓	$3 \times 10^{-4}$ s	7.79 — 0.04%	—
Lead-212 (Thorium B) ↓	10.6 h	—	0.12 — 0% 0.24 — $\sim 36\%$ 0.30 — $\sim 3\%$
Bismuth-212 (Thorium C) ↓	60.5 m	6.04 — 25% 6.08 — 10% others — 1%	0.04 — $\sim 0\%$ others — very weak ----- 0.73 } most 0.79 } abundant 1.08 } $\sim 10\%$ 1.62 }
64% 36% ↓ Polonium-212 (Thorium C') ↓	$3 \times 10^{-7}$ s	8.78 — $\sim 64\%$ others of higher energy—very weak	—
Thallium-208 (Thorium C') ↓	3.1 m	—	0.28 — $\sim 3\%$ 0.51 — $\sim 8\%$ 0.58 — 31% 0.86 — 4% 2.62 — 36%
Lead-208 (Thorium D)	Stable	—	—

Figure 1. Thorium - (4n) series starting at thorium-228.

standards were calibrated by comparing their  $\gamma$ -ray emission rates, in a reproducible geometry, with that of an NBS working standard of thorium-228 which was prepared by plating some thorium-228 onto a platinum disc, and then flaming it to red heat. This provides an ideal source for absolute  $\alpha$ -particle counting, upon which the calibration of the working standard was based.

Since each decay of thorium-228 leads to precisely 5  $\alpha$  particles and the 2.614 MeV  $\gamma$ -ray abundance is known<sup>(2)</sup>, the equilibrium  $\gamma$ -ray emission rate can be determined.

In order to reduce radon escape during calibration of the working standard, it was covered with thin mylar films which resulted in  $\alpha$ -particle-energy absorption in the films and limited the amount of film that could be used. For example, when 60- $\mu$ m-thick mylar was used as a covering layer on a polonium-210 source, the energy degradation and  $\alpha$ -particle scattering caused losses of the order of 1% in the counting rate, which was unacceptable. Counting rates, with 2 covering layers of 4- $\mu$ m-thick mylar over a polonium-210 source, were checked in the  $1\pi\alpha$  chamber and there was no loss in count rate due to the mylar within  $\pm 0.2\%$ . We used 2 or 3 layers of this 4- $\mu$ m mylar in order to minimize the probability of a direct escape path for radon atoms from the source. The source was covered, placed in a vacuum chamber, and allowed to come to equilibrium, which was checked by monitoring the count rate of the 8.78-MeV polonium-212  $\alpha$  particles, using a surface-barrier silicon detector (Figures 2 and 3). From observation of a background with a 10-hour half-life immediately after a source had been withdrawn from the chamber, we concluded that some radon atoms were still escaping from the source.

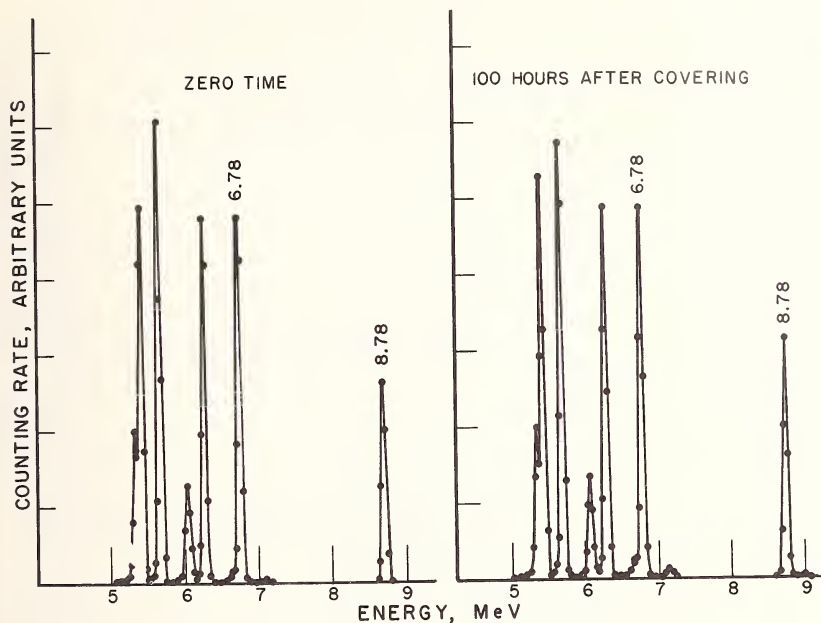


Figure 2.  $\alpha$ -particle spectra of a thorium-228 source immediately after covering the source with thin plastic and 100 hours after (see text). The highest energy peak, 8.78 MeV, has increased significantly over the initial value indicating accumulation of progeny of radon-220 behind the mylar covering layer.

The problem, then, was to determine how much radon was escaping. We counted the source for  $\alpha$ -particle activity, then took it out of the counter, placed some very thick plastic tape over the source and within, say, five minutes, started counting the source for  $\gamma$ -ray count rate above 84 keV with a NaI(Tl) detector. Since some radon had been escaping from the source, the daughters following radon were not in equilibrium with the thorium. We followed the  $\gamma$ -ray count rate of the heavily-taped

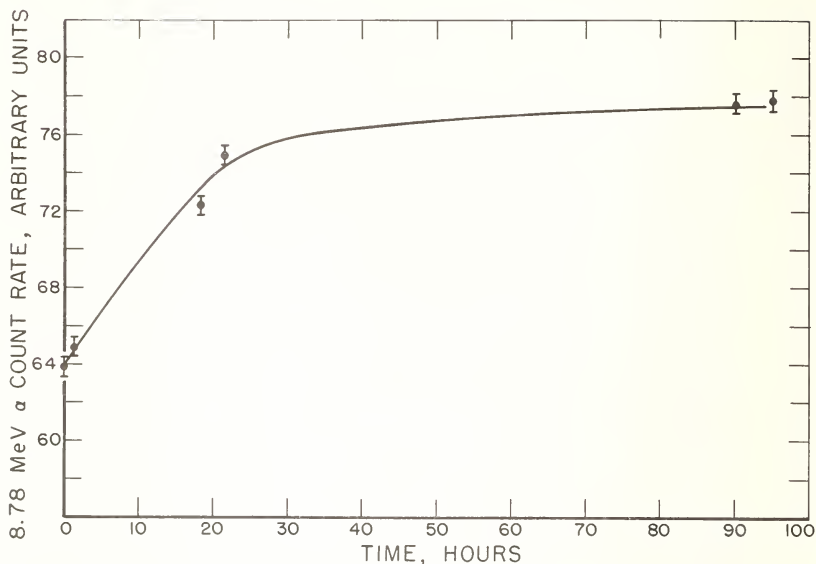


Figure 3. 8.78-MeV  $\alpha$ -particle counting rate for a thorium-228 source, as a function of time, using excessively-porous mylar covering layer (not included among points shown in Figure 5).

source over a period of time, and found the initial rate increased and finally came to equilibrium after 3 or 4 days (Figure 4), this period having been determined by 10.6-hour lead-212. We repeated this experiment several times. Each time, after measuring the  $\alpha$ -particle and  $\gamma$ -ray count rates, we removed all the heavy and thin tape, recovered the source with new thin tape, and after a new "equilibrium"  $\alpha$ -particle count rate was obtained, remeasured the source in the same geometry counter as before, following which, the source was re-encapsulated with the heavy tape and  $\gamma$ -ray counted again.

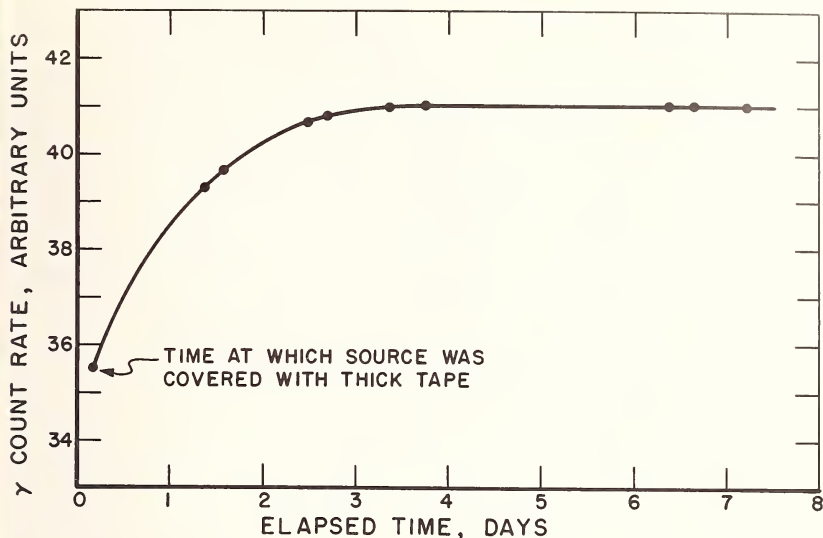


Figure 4.  $\gamma$ -ray count rate for a thorium-228 source as function of time after thick plastic encapsulation.

Figure 5 (curve B) shows the result of 3 sets of counts. The ordinate of figure 5 is the thorium-228 source count rate with only the thin mylar-film coverings. As the porosity of the thin mylar film is a random property, there were different amounts of radon escaping during the three experiments. The abscissa, labelled  $\Delta\gamma$ , is the difference (in percent) between the initial  $\gamma$ -ray count rate immediately after encapsulating the source and the final equilibrium count rate. Extrapolating to  $\Delta\gamma=0$  (the condition for zero radon loss) gives a value, which, when normalized by the vacuum-chamber-surface barrier-detector geometry factor, gives the  $\alpha$ -particle activity for

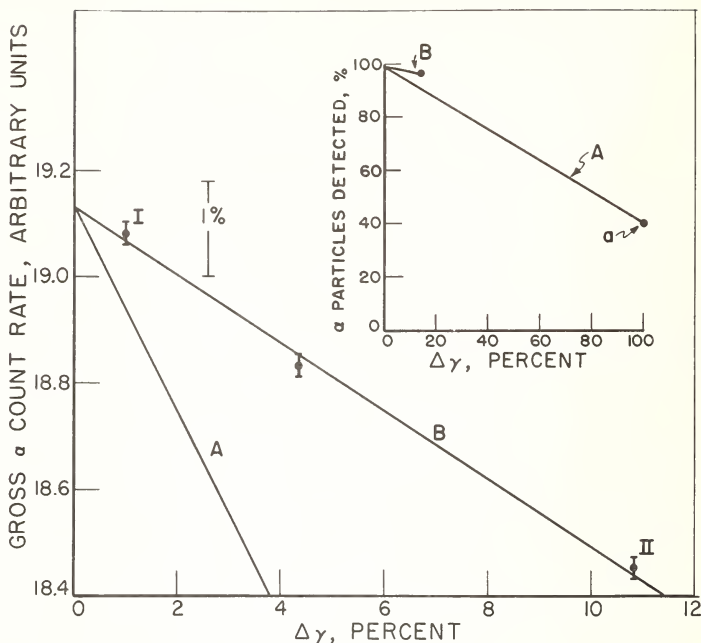


Figure 5.  $\alpha$ -particle counting rate of thorium-228 source under three different conditions of radon escape (curve B) as a function of  $\Delta\gamma$  (defined in text). Escape is least for condition 1, greatest for 11. Inset shows curve A for  $\Delta\gamma$  from 0 to 100%.

thorium-228 in equilibrium with its daughters. This number then is divided by 5 to give the activity of bismuth-212, and multiplied by  $0.3600 \pm 0.0003$ , <sup>(2)</sup> the number of 2.614-MeV  $\gamma$  rays per decay of bismuth-212.

A few explanatory words are in order with reference to Figure 5. Of the five  $\alpha$ -particle decays per decay of thorium-228 (cf Figure 1), two occur before radon 220, and, 95% of the  $\gamma$  rays above 240 keV occur in the progeny of radon-220. Note,

that if all the radon escapes the source, (condition at point a) the source would still emit 2 of the 5  $\alpha$  particles, but essentially none of the  $\gamma$  rays. The curve actually obtained (B) has a slope about half that of A, due to detection of  $\alpha$  particles from some of the decaying radon atoms (and/or their daughters) which had escaped the source. Calculations show that this detection efficiency amounted to about 0.75 percent of these "external" decays, and the fact that the experimental values fall on a straight line show that this efficiency was constant for the three source conditions.

One other interesting standard developed this year which is now available is that of cadmium-109. Cadmium-109 decays by electron capture. It has an 88-keV  $\gamma$  ray <sup>(3)</sup> of approximately 4% abundance. One might calibrate this radionuclide for total activity by conversion-electron-x-ray coincidence counting, but we were interested only in the  $\gamma$ -ray emission rate. In Table 1, our  $\gamma$ -ray point sources are listed in the center column, and as we are trying to cover as large a range of energies as possible, and inasmuch as the  $\gamma$ -ray abundance of cadmium-109 is not known very accurately, an activity calibration would not have been as valuable for our purposes, as the  $\gamma$ -ray emission rate. The calibration was accomplished with the use of a  $4\pi\gamma$ -sodium iodide well crystal. Corrections had to be determined for some eight effects, and this work will be described in a forthcoming publication.

Recognizing the importance of impurities in standards and the troubles that can arise from their presence, we have been looking with improved capability at the impurity levels since we acquired several lithium-drifted germanium detectors. Whereas in previous years we could easily have had small amounts of  $\gamma$ -ray impurities which could not be detected by conventional sodium-iodide  $\gamma$ -ray spectrometry, their presence can now be very easily seen with a germanium detector

and we go to considerable pains and effort to remove the impurities.

The point that was brought up a few minutes ago by Dr. McNair is a very important one, and it emphasizes the fact that the user of a radioactivity standard should be well aware of not only the purpose for which he wants it but what actually is in the standard. To this end, we not only try to determine the level of impurity, but we include this information in our certificates and also indicate the method used to determine such levels.

Another interesting experiment which was not directly related to  $\alpha$  standardization but which illustrates an elegant technique for nuclear-instrument calibration, was associated with the measurement of the half-life of the 74-keV state of neptunium-239 arising in the decay of americium-243. <sup>(4)</sup> In order to measure very short half-lives (this one turned out to be  $1.38 \pm 0.03$  nanoseconds), a time-to-pulse-height converter is customarily used in conjunction with a pulse-height analyzer, the latter being calibrated in terms of time-per-channel. Conventionally, this calibration is accomplished by insertion, into the electronics system, of different lengths of coaxial cable of known time delay. This method is frequently subject to systematic errors arising from subtle effects due to types of connectors and fittings used with the cables. In this experiment, the calibration was accomplished without disturbing any of the cables in the instrumentation. A universal standard available to everyone - at no cost -, the speed of light, was used. By increasing the distance between a pulsed light source and a phototube, and measuring the corresponding times of flight, the system was readily and accurately calibrated.



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A. Spernol

Would Mr. Garfinkel be so kind and inform us if any of your niobium-93m is available, as we are very much interested in pure niobium-93m.

S. B. Garfinkel

We hope to do it. We haven't done it yet. If you want some niobium-93m now, we have some, but it's in the presence of niobium-94!

W. B. Mann

Thank you very much, Mr. Garfinkel.

May I make one more comment perhaps on Dr. Rytz's previous statement, and Dr. Caswell's expressing surprise that there will be no more intercomparisons. I forgot to mention at that point that we were asked with NRC Ottawa, Dr. Baerg, to look into the question of trying to do tracer intercomparisons of carbon-14 using possibly niobium-95 oxalate which has an end-point energy of the beta spectrum about the same as carbon-14. We were asked to do this at the November meeting in Sévres last year. I suppose you consider this to be still on the program, Dr. Rytz?

A. Rytz

Yes.

W. B. Mann

It may be difficult, Dr. Rodriguez-Pasqués, Miss Cavallo, Dr. Landgrebe, and Mr. Garfinkel are all looking into this, and I am too, with Dr. Baerg. So this is still on the program apparently. But it is so difficult that I'm not quite sure that we are going to find a solution in the near future. I think most carbon-14 standards are unfortunately compared with the National Bureau of Standards' standards and this is rather an invidious position and we'd rather have an international comparison on this as soon as possible. I hope that the participants this afternoon will not mind perhaps letting us have the manuscripts so we can make some effort to publish these proceedings. I think it would be very interesting. I must say it has been a great privilege to have so many distinguished people here today and talking. It has been a great pleasure and I wish to thank you all.





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